Emerging technologies for the recovery of rare earth elements (REEs) from the end-of-life electronic wastes: A review on progress, challenges, and perspectives

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REVIEW ARTICLE

Emerging technologies for the recovery of rare earth elements (REEs) from the end-of-life electronic wastes: a review on progress, challenges, and perspectives

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Abstract

The demand for rare earth elements (REEs) has significantly increased due to their indispensable uses in integrated circuits of modern technology. However, due to the extensive use of high-tech applications in our daily life and the depletion of their primary ores, REE's recovery from secondary sources is today needed. REEs have now attracted attention to policymakers and scientists to develop novel recovery technologies for materials' supply sustainability. This paper summarizes the recent progress for the recovery of REEs using various emerging technologies such as bioleaching, biosorption, cryo-milling, electrochemical processes and nanomaterials, siderophores, hydrometallurgy, pyrometallurgy, and supercritical CO_2. The challenges facing this recovery are discussed comprehensively and some possible improvements are presented. This work also highlights the economic and engineering aspects of the recovery of REE from waste electrical and electronic equipment (WEEE). Finally, this review suggests that greener and low chemical consuming technologies, such as siderophores and electrochemical processes, are promising for the recovery of REEs present in small quantities. These technologies present also a potential for large-scale application.

Keywords Biosorption · Bio-sorbent · Desorption · Rare earth metals · Recovery

Introduction

Rare earth elements (REEs) form a group of 17 metals naturally found in the environment, including 15 lanthanides, scandium, and yttrium (Mikołajczak et al. 2017). They can be divided into light rare earth, which includes cerium (Ce), lanthanum (La), neodymium (Nd), praseodymium (Pr), samarium (Sm), and heavy rare earth, comprising dysprosium (Dy), erbium (Er), europium (Eu), gadolinium (Gd), holmium (Ho), lutetium (Lu), terbium (Tb), thulium (Tm), ytterbium (Yb), and yttrium (Y). REEs can be extracted from reserves of bastnaesite, monazite, xenotime, and ion adsorption clays (Wiche et al. 2017). However, their presence in the environment commonly makes mining challenges (Gambogi 2019). Therefore, despite their relative abundance in the earth's crust, many of them are in supply risk. The European Commission placed REEs in the European list of critical raw 42 materials, due to economic importance and low availability (EU 2014). In 2017, the European Commission published a new report to classify the critical elements according to their economic importance and supply risk (EU, COM- 2017). In 2018, the USA also added REEs to the list of 35 critical minerals (US Department of Interior 2018).

According to Gambogi (2019), the biggest producer of REEs is China. In 2018, the US net import reliance on rare earth compounds was 100% (80% from China), summing up to US\$160 million. Prices highly vary depending on the element. For instance, for Ce_2O_3, it is around US\$1500 per ton,

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while for Sc_2O_3, it can reach up to US$3,000,000 (Liu et al. 2018).

REEs are widespread through diverse applications in our daily life. Thanks to their unique physicochemical properties, REEs can be applied in different fields such as metallurgy, optical/magnetic/chemical engineering, high-temperature superconductors, laser materials, secondary batteries, and luminescence (Banda et al. 2012; Charalampides et al. 2015). They are used for manufacturing different products, such as catalysts, magnets, alloys, and phosphors (Swain and Mishra 2019). REEs also provide colors to liquid crystal displays and cathode ray tubes (Menad and Houwelingen 2015; Sahan et al. 2019). They can have different applications in metallurgy such as, in alloying and purification of materials, a metamorphosis of inclusions and refinement of microstructure (Das and Das 2013; Emmanuel et al. 2012; Jorjani and Shahbazi 2016; Xiong et al. 2009).

Although the irrefutable economic importance of REEs, they are usually lost as wastes, as a consequence of the linear flow of goods in the economy. Being commonly used materials in high technology, REEs' contents in waste of electrical and electronic equipment (abbreviated as WEEE or e-waste) are considerably high (Menad and Seron 2016).

Globally, the waste generated from electrical and electronic equipment (WEEE) has increased dramatically. According to Balde et al. 2015), global WEEE was generated at around 50 million tonnes in 2018. The increasing burden of WEEE has tangible environmental and social concerns. Unsafe WEEE disposal can lead to many health implications due to their hazardous nature, being composed of substances such as chlorofluorocarbons and polybrominated diphenyl ethers (Commission, E 2008). Besides, a huge amount of valuable materials is being wasted globally since WEEE can act as a secondary resource for critical and precious elements. They contain a significant amount of critical base metals and technology metals (Rene et al. 2017; Robinson 2009). Despite these facts, only ~ 1% of the REE is recycled from end-products, with the rest deporting to waste and being removed from the materials cycle (Jowitt et al. 2018; Dodson et al. 2012). This opens a new chapter in the recovery of critical and precious elements from a different end-of-life electronic wastes; in addition to this, the manufacturing technology for the recovery of metals depends upon these imported rare earth elements such as neodymium, lanthanum, yttrium, and cerium, which account for about 85% of the global production but recovery of these metals has some limitations due to technical problems (Işıldar et al. 2018). Separating these REE materials from the products in a tiny amount still poses challenges (Du and Graedel 2011). The most common methods used to recover REEs from WEEE include mechanical treatments, at a pre-processing stage, and metallurgical treatments as main processes for metals refining. Pyro- and hydrometallurgical technologies are energy and resource intensive and usually address only the recovery of precious and base metals (Bigum et al. 2012). Also, dust from dismantling and grinding WEEE are usually not recycled, being currently landfilled (Marra et al. 2018), even if the highest contents of REE during mechanical treatment of WEEE are found in the collected dust (Oguchi et al. 2012).

In the past 10 years, more research was carried out to recover REEs from a different end-of-life electronic wastes using conventional approaches such as precipitation, filtration, pyrometallurgy, hydrometallurgy, liquid-liquid solvent extraction, among others (Sethurajan et al. 2019). For instance, Marques et al. (2013) studied the recovery of critical elements from discarded printed circuit boards (PCB) using pyrometallurgical and hydrometallurgical methods. They showed that PCB had a composition of 26.5% plastic, 16.5% non-ferrous metals, 38.1% ferrous metals, and 18.9% others. Besides, they proved that the concentration of metals in waste printed circuit boards is higher than that of the ores of the natural minerals (Marques et al. 2013). Still, these conventional approaches are considered as promising technologies for the recovery of REEs. However, these approaches cannot extract or recover the REEs with low concentrations and have a low affinity towards targeted metal. High consumption of reagents and energy, generation of secondary metabolites, and lack of economic advantages restrict their efficiency for the recovery of metals from a different end-of-life electronic wastes (Attallah et al. 2016; Larsson and Binnemans 2017; Marra et al. 2018; Parhi 2012; Qi et al. 2017; Tunsu and Retegan 2016; Wang et al. 2010; Wu et al. 2018; Xiong 2008; Xiong et al. 2008; Yunnen et al. 2015; Zheng and Xiong 2011). Therefore, there is a need to develop and design cost-effective and eco-friendly methods to recover metals from different end-of-life electronic wastes, considering the problems mentioned above. This review discusses comprehensively recent technologies and future challenges and perspectives for the efficient recovery of rare earth elements from secondary sources, including different end-of-life electronic wastes. It also summarizes the engineering and economic point of view of both conventional and innovative separation and recovery technologies for WEEE with special attention being given to the overall sustainability.

Problems and challenges in recycling electronic wastes

One of the significant challenges in the recycling of electronic waste is the very low quantity of REEs in each device. This may create the wrong idea that recovering REEs from WEEE is not profitable. The recovery efficiency is low due to the lack of an appropriate design (Wang et al. 2016). Profound research carried out in the past few decades for the recovery of REEs from electronic wastes (e.g., fluorescent lamps, magnets, NiMH batteries, mobile phones, and others) shows that about 99% of REEs are recycled (Darnerud 2008; Marinković et al. 2010; Abdelouahab et al. 2011; Zhang et al. 2015; Cucchiella et al. 2016; Furberg et al. 2019; Hsu et al. 2019; Meng et al. 2019; Yoshida and Terazono 2010).

Kell (2009) recovered REEs such as Nd, Pr, and Dy without the involvement of non-REE from industrial scrap magnets and industrial NdFeB magnets using membrane-assisted solvent extraction in 120-h run. Similar results have been reported by Khaliq et al. (2014) for electronic wastes. Some electrical and chemical companies are integrated to develop procedures and processes for the recovery of REEs having high purity and efficiency from electronic waste. It has also been reported that this mature process has impressive advantages such as low environmental footprint, which enhances shorter lead time as well as a cheaper source when compared with the primary production of the electronic material (Stevens and Goosey 2008).

Most of these approaches can be applied for the recycling of electronic wastes by using sorbents for solid-phase extraction or liquid-liquid extraction processes in a column or batch system (Jiang et al. 2012). Moreover, the recovery of these elements from electronic wastes, such as metal alloys in magnets, need special treatment than the above approaches. There is also a need to develop products recycling policy and other networks that can assure quality, price efficiency of the separation, and recovery of REEs. It is also essential to develop strategies and for preserving REEs resources (Hanafi et al. 2012; Jiang et al. 2012; Yao et al. 2018). In general, to develop novel and sustainable recycling techniques, some challenges need to be addressed in the future, to implement REEs recovery on a large scale (Dalrymple et al. 2007).

Recycling of electronic waste

Electronic wastes are complex because they contain oxides, metals, non-metals, and polymeric materials, rendering their separation and recovery very challenging. Details about WEEE recycling technologies are presented and discussed below.

Pre-treatment of electronic waste

Pre-treatment is one of the first steps during WEEE recycling, involving disassembly or dismantling, physical separation, and size reduction. During the recovery of metals and other REEs from electronic waste, selecting an appropriate technology is challenging because these elements differ in their type, metal content, and composition (Cui and Forssberg 2003; Yazici and Deveci 2009; Yazici et al. 2015; Goosey and Goosey 2019). Thus, it is necessary to enhance the technical efficiency of the recovery process with a material-centered approach as well as to reduce its pollution (Yazici and Deveci 2009; Priya and Hait 2018; Yamane et al. 2011).

One of the downsides of currently used pre-treatment technologies is the loss of critical elements, for instance, as shredding dust (Sethurajan et al. 2019). Thus, optimization of the pre-treatment stage is still needed to avoid the loss of the REEs during the recovery process (Oguchi et al. 2011). It is also necessary to integrate the pre-treatment process with chemical and other physical methods for the energy recovery and removal of hazardous components (Marra et al. 2018) as shown in Fig. 1.

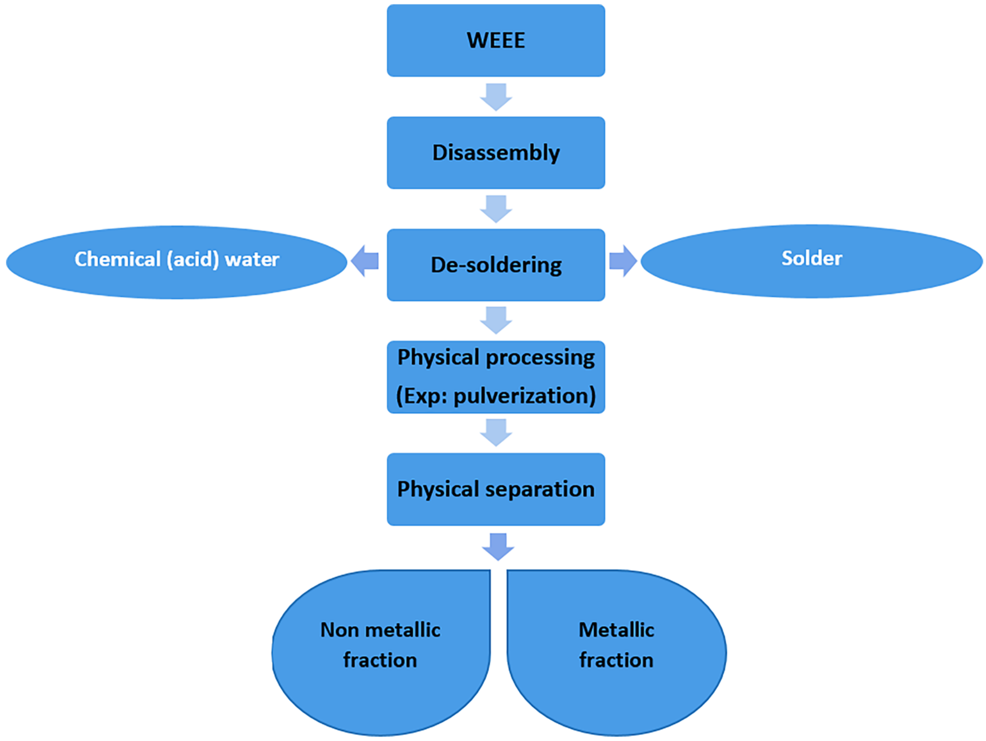
Dismantling/manual disassembly is the process in which hazardous material and other streams are removed from the components of electronic wastes such as screens, batteries, capacitors, PCB, computers CPU and RAM for the selective recovery of the REEs, and other high-value products (Lee et al. 2004). Moreover, in terms of process view, it allows separating the metallic parts from the non-metallic elements, such as ceramics and plastics, to increase the overall potential of recycling in a circular economy context (Li et al. 2007). However, there are some difficulties in separating and recovering REEs from the different types of electronic waste due to high costs of automation for the specific application, resulting in manual dismantling, which makes this stage labor intensive (Kopacek 2016; Lindkvist et al. 2017; Park et al. 2015). To overcome this problem, there is an approach developed by Wang et al. (2012) called "Best-of two-Worlds" (Bo2W) which states that it is necessary to ingrate the manual sorting or dismantling with the automatic developed process for the efficient recovery technologies of the REEs from the electronic waste (Wang et al. 2012).

Mechanical pre-treatment process

The second step in the recovery of metals and other REEs from electronic waste is mechanical treatment. In this step, metals and other value products are separated and liberated using physical processes. They can be segregated by shape, size, weight, density, and magnetic and electrical characteristics to improve the technical and economic aspects of the recovery process (De Oliveira et al. 2012). For instance, Kaya (2016) investigated the recovery of metals from waste printed circuit boards using physical and chemical processes. The researcher showed that hammering and shredding electronic waste reduced it to a suitable size, enhancing the efficiency of the recovery process (Kaya 2016). However, recent research by Martino et al. (2017) showed that reduction of the size and liberation using electrodynamic fragmentation is an unconventional process for the recovery of the metal due to fine grinding (typically < 200 mm) of the electronic waste (Martino et al. 2017).

Other researchers (Ogunniyi et al. 2009; Yazici and Deveci 2009; Yazıcı et al. 2010) have also reported a similar finding to recover metals from PCB. These studies show that if the size of the electronic waste is < 75 mm, it leads to trouble in

Fig. 1 Integrated pre-treatment methodology for WEEE



separation and liberation. Moreover, they claimed that getting the required amount of the size reduction of the electronic waste can increase the liberation as well as the physical separation of the metals from the electronic waste, before applying different extraction technologies.

The main purpose of physical separation is to segregate the materials and increase the benefit of WEEE based on their size, at a low price. Physical separation is based on differences in conductivity, specific gravity, brittleness, hydrophobicity of the phases, and magnetic susceptibility (Veit et al. 2005; Wills and Finch 2015; Yazici and Deveci 2015). Magnetic separation, for instance, can be applied to remove and separate ferrous metals from electronic waste, as a magnetic fraction as shown in Fig. 2 (Pant et al. 2012; Yazici and Deveci 2009). Air classification can be used to separate fine plastics or fluffy material (Lee et al. 2004; Zhao et al. 2012; Zhou and Qiu 2010).

Moreover, light metals which have high conductivity to density ratio can also be separated from heavy non-ferrous metals and non-metallic fraction using eddy current. In this way, they are recovered based on their conductivity using the electrostatic separation method (Taurino et al. 2010). In general, the metals and REEs found in the electronic waste can be separated using the difference in density as well as specific gravity separation methods such as jigging, media separation, and shaking tables (Ongondo et al. 2011). However, the effectiveness of the physical process may decline with decreasing in size and with the different physical properties of the fractions (Hageluken 2006; Hagelüken 2006; Kamberović et al. 2018; Yazici and Deveci 2014; Zhao et al. 2004). Some physical separation approaches for the recovery of metals from WEEE are shown in Table 1.

There is dire need to conduct more experimentation to increase the recovery of metals with low impurities from electronic waste and to assess the fate of the critical elements in the future (Duan et al. 2009; Galbraith and Devereux 2002; Guo et al. 2011; Kumar et al. 2018; Ogunniyi and Vermaak 2009a; b). Duflou et al. (2008) show that automated disassembly is not technically and economically feasible. Moreover, they claimed that using a general standard procedure for grinding electronic waste can lead to loss of precious metals, which cannot be recovered by refining or downstream process. They recommended that the optimization of the manual disassembly process is needed during the pre-treatment (Duflou et al. 2008; Buchert et al. 2012). In another study by Zhang and Xu (2016), a mechanochemical method was used as pre-treatment for the recovery of metals. This method has higher efficiency than direct hydrometallurgical technology and decreases the loss of precious elements during the process. Recycling of electronic waste for the recovery of precious metals must be more product oriented than a material-centered approach.

Recovery and separation of REE from electronic waste

The prime interest for the recycling of electronic wastes is the recovery of REEs. The processes used in REEs recovery and separation are bioleaching, biosorption, siderophores,

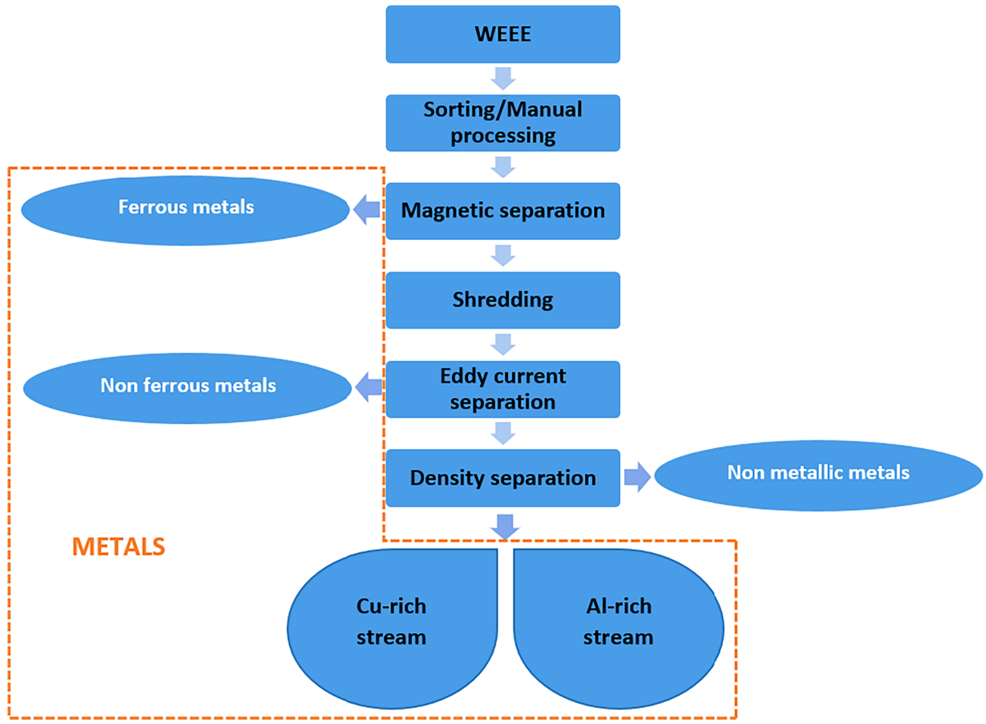


Fig. 2 Physical separation scheme for separation of non-metallic fractions from metals

hydrometallurgy, pyrometallurgy, and carbon-based material. All methods and procedures have some advantages and disadvantages. In this section, different techniques and mechanisms used for the recovery of REE from electronic waste are discussed.

Bioleaching

Bioleaching is an effective technology of metal recovery from primary and secondary sources using microorganisms such as extremophiles, moderately thermophilic bacteria, and mesophiles.

It is widely used in commercial recovery of metals especially molybdenum (Mo), copper (Cu), zinc (Zn), nickel (Ni), arsenic (As), cobalt (Co), antimony (Sb), gallium (Ga), palladium (Pd), platinum (Pt), and osmium (Os) from their ores (Brierley and Brierley 2013; Schlesinger and Sole 2011). Bioleaching can also be applied for the extraction of metals from electronic waste, mine, flay ashes, contaminated soils, sludge, and spent catalysts (Ishigaki et al. 2005; Liu et al. 2007; Liu et al. 2008; Ming et al. 2012; Chang et al. 2014; Potysz et al. 2016; Deng et al. 2013; Lee and Pandey 2012; Yin et al. 2014).

Johnston et al. (2013) investigated the extraction of REEs from electronic waste using bio-recovery, which includes bioreduction, acidolysis, biomineralization, and cyanogenic bioleaching. The results showed that when compared with the other processes such as hydrometallurgy and pyrometallurgy, bio-recovery is a more cost-effective and environment-friendly process to recover valuable metals from WEEE. These technologies have opened the door to develop a new understanding of biotechnological process optimization, metal mobilization, and toxicity mechanisms for the recovery of REEs from electronic waste. Ilyas and Lee (2014) have conferred about 99% copper metal recovery from biomass waste using bioleaching. They found out that copper metal recovery

Table 1 Some physical separation approaches for the recovery of metals from electronic waste

Approaches	Targeted material	Particle size (mm)	Reference
Electrostatic using electrical conductivity	Non-metals from metals	0.2–5	Zhang et al. (2017)
Magnetic using magnetic susceptibility	Non-ferrous metals and non-metals (para-/dia-magnetics) from metals	< 5	Zhang et al. (2017)
Flotation using surface properties	Metals from Non-metals (hydrophobic)	0.075–1	Gallegos-Acevedo et al. (2014)
Gravity using specific gravity	Non-metals from metals	0.05–10	Veit et al. (2014)
Eddy current using electrical conductivity or specific gravity	Light metals from non-conductive and non-conductive materials	> 5	Yazıcı et al. (2010)

from biomass generates fewer contaminants in comparison with conventional metal processing. Moreover, they claimed that this process could also be used for the recovery of metals from their ores and other secondary sources such as electronic waste. Some studies on REE extraction from WEEE through bioleaching are outlined in Table 2.

Biosorption

Biosorption is one of the modern biological methods that get the most attention in the field of recovery of metals from electronic waste, due to unique properties such as high recovery efficiency for metals in low concentration, high regeneration, fast kinetics, and non-generation of secondary residues. It is cost-effective, can be efficiently operated and used in situ, shows high efficiency in the removal of contaminants from aqueous solution, and does not produce any chemical sludge. Moreover, it can be easily integrated with any system as compared with the conventional methods (Das et al. 2008; Liu et al. 2016a; Singha and Das 2011; Xiong et al. 2012a, b; Xiong et al. 2013).

In the past decades, extensive studies have been carried out for the recovery of rare earth metals from electronic waste using bacteria, fungi, and algae as biosorption material (Diniz and Volesky 2005; Kazy et al. 2006; Palmieri et al. 2002; Shuxia et al. 2011; Vijayaraghavan and Jegan 2015). These researchers have investigated the recovery of lanthanum from electronic waste by using *Sargassum biomass*, *Pseudomonas* sp., and *Agrobacterium* sp. HN1 as an adsorbent. For biosorption of neodymium, they have used *Monoraphidium* sp., bakers' yeast, *Penicillium* sp., *Saccharomyces cerevisiae*, *Kluyveromyces marxiamus*, *Candida colliculosa*, and *Debaryomyces hansenii* (Anagnostopoulos and Symeopoulos 2013; Palmieri et al. 2000). Sert et al. (2008), Xiong et al. 2008; Oliveira et al. 2011 and Shuxia et al. (2011) have successfully reported biosorption of cerium by *Platanus orientalis* leaf powder and *Agrobacterium* sp. *HN1*. These approaches are found highly efficient and cost-effective for the recovery of metals from solutions. Moreover, biosorption of metals depends upon temperature, pH, agitation rate, contact period, and initial metal concentration, as shown in Fig. 3. The compatibility of the adsorbent's technology and optimized process parameters also affect recovery efficiency of metals from electronic waste (Awwad et al. 2010; Chakravarty and Banerjee 2012; Chen et al. 2011; Hu et al. 2004; Romera et al. 2007; Zheng and Xiong 2011; Kazy et al. 2006; Kütahyali et al. 2010; Qing 2010; Shuxia et al. 2011; Xiong et al. 2011; Birungi and Chirwa 2014). In general, some of the studies on REEs extraction from WEEE through biosorption are outlined in Table 3.

Biosorption technology has shown to be a promising and low-cost technique for the recovery of REEs from electronic waste. However, further studies must be carried out to identifying the exact compatibility of various bio-sorbents for the recovery of metals. There is a need to develop a method for the

Table 2 Recovery of REEs by bioleaching from WEEE

Microorganism	Targeted elements	Efficiency	Reference
Sulfobacillus thermosulfidooxidans	Cu, Ni, Zn	89%, 81%, 83%	Ilyas et al. (2007)
Gallionella sp.	Cu	95%	Oguchi et al. (2012)
At. thiooxidans	Cu, Ni, Zn	94%, 89%, 90%	Liang et al. (2010)
Acidophilic consortium	Cu, Al, Zn	97%, 88%, 92%	Hussein et al. (2004)
Leptospirillum ferrooxidans	Cu	95%	Bas et al. (2013)
Acidithiobacillus thiooxidans	Cu	98%	Hong and Valix, 2014)
Ferroplasma acidiphilum, *Sulfobacillus benefaciens*, *At. caldus*, *L. ferriphilum*	Cu	99%	Bryan et al. (2015)
Penicillium simplicissimum, *Aspergillus niger*	Cu, Al, Ni, Zn	65%, 95%, 95%, 95%	Brandl et al. (2001)
Pseudomonas plecoglossicida, *Pseudomonas fluorescens*, *Chromobacterium violaceum*	Au	69%	Brandl et al. (2008)
P. chlororaphis	Au, Ag, Cu	8%, 12%, 52%	Gao et al. (2017)
Thermoplasma acidophilum, *Sb. thermosulfidooxidans*	Cu, Zn, Ni, Al	86%, 80%, 74%, 64%	Ilyas et al. (2010)
Sb. thermosulfidooxidans	Cu Al, Zn, Ni	95%, 91%, 96%, 94%	Ilyas and Lee, 2014)
At. ferrooxidans	Cu	95%	Chen et al. (2015)

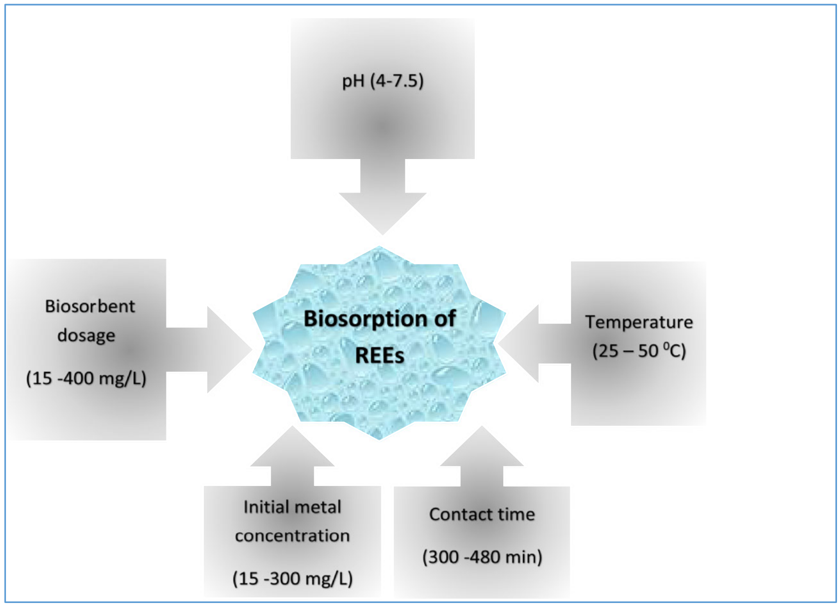


Fig. 3 Important parameters and their ranges that affect the biosorption of REEs

regeneration of biosorption material into recovered metals that can be sold in the market.

Siderophores

Siderophores are small, high-affinity iron-chelating compounds secreted by microorganisms such as bacteria, fungi, and grasses. The chelator molecules are produced to hunt Fe^{+3} from the environment to satisfy the microorganism's metabolic needs. They are known as the best ligands for ferric ions (Chaturvedi et al. 2012; Hider and Kong 2010). Besides, siderophores may also have a strong affinity towards other metals, allowing a new understanding of an interesting biological and eco-friendly method that can be used for the recovery of REEs. Many researchers have compared the REEs recovery efficiency and chemical leaching of siderophores (Bau et al. 2013; Christenson and Schijf 2011; Mohwinkel et al. 2014). They have shown that siderophores have a high affinity towards desferrioxamine, which is abundant naturally. Mohwinkel et al. (2014) have also experimented with the recovery of REEs using siderophores of desferrioxamine.

Table 3 Recovery of REEs by biosorption from WEEE

Bio-sorbent material	Targeted material	Efficiency (mg/g)	Reference
Bacillus cereus	Ag^{+}	91.75	Li et al. (2011)
Klebsiella sp. *3S1*		141.1	Muñoz et al. (2017)
Chemically modified chitosan resin		413.62	Donia et al. (2007)
Thiourea modified alginate	Au^{3+}	1668.25	Gao et al. (2017)
Racomitrium lanuginosum		37.2	Sarı and Tuzen (2009)
Bayberry tannin	Pd^{2+}	33.4	Kim and Nakano 2005)
Pleurotus ostreatus basidiocarps	Y^{3+}	54.54	Hussien (2014)
Prawn carapace	La^{3+}	1000.00	Das et al. (2014)
Chlorella vulgaris		74.60	Birungi and Chirwa 2014)
Sargassum sp.		91.68	Birungi and Chirwa (2014))
Platanus orientalis	Ce^{3+}	32.05	Heilmann et al. (2015)
Green seaweed (*Ulva lactuca*)		69.75	Vijayaraghavan (2015)
Free *Turbinaria conoides* (brown seaweed)		146.4	Vijayaraghavan (2015)
Chlorella vulgaris	Pr^{3+}	157.21	Kücüker et al. (2016)
Sargassum sp.		98.63	Vijayaraghavan (2015)

Researchers have found higher recovery efficiency for lithium and molybdenum, and lower recovery efficiency for cerium, mainly due to the cerium complex formation with siderophores, which was inhabited by the competing cations present in the solution, such as Fe^{+3}.

The recovery of REEs using siderophores is cost-effective, rapid, reversible, and eco-friendly technology compared with conventional methods for the recovery of REEs which are available in small concentration from a different end-of-life electronic wastes (Christenson and Schijf 2011; Hernlem et al. 1996; Hernlem et al. 1999; Hider and Kong 2010; Su et al. 2011). Siderophores have high stability to enhance the recovery of REE from secondary sources, as the different end-of-life electronic wastes. It has a unique application for the recovery of REEs in the future and can be applied in a vast environmental field.

Pyrometallurgy

Pyrometallurgy is one of the known thermal treatments that can be used for the recovery of metals from electronic waste. This process includes smelting in a plasma arc furnace, blast furnace or copper smelter, incineration, and high heat roasting in the presence of selective gases to recover mainly non-ferrous metals. Moreover, this process is characterized by high energy consumption. However, it has high efficiency for the recovery of selective REEs from electronic wastes. Cui and Roven (2011), as well as Hsu et al. (2019), reported about 70% recovery of REEs from electronic wastes such as PCBs using this process. Bernardes et al. (2004) showed that the smelting process could accept any feedstock of electronic wastes as raw material for the recovery of copper and other precious metals (Bernardes et al. 2004).

Cui and Zhang 2008a, b) also reported a similar result for the recovery of Cu and other precious metals from electronic waste such as PCBs using smelting integrated with vacuum metallurgy separation. They have shown that PCBs can produce a mixed oxide (mainly Pb and Zn), slag by a top blown reactor, and an alloy of Cu-Ni-Si and observed that high pressure enhanced the separation of Bi, Sb, Pb, and other heavy metals. However, this process has some limitations on the pillars of the sustainability goal of the UN. One is related to the environmental problem associated with the release of toxic materials, the formation of dioxin, and high state of the art smelting, which can lead to the creation of slags and another industrial system. Another issue consists of the need for a more advanced control emission system, which is quite more costly (Cui and Zhang, 2008a, b). Another technical problem is that this process can recover only Cu. Still, it cannot recover other metals such as Al and Fe and results in higher losses of precious and base metals as metals. Hence, metals can be concentrated in the slag, leading to less efficiency. Besides, more time in separation and recovery is needed for the metals which have high economic value. So, there is a need to integrate the pyrometallurgy with the electrorefining process for complete metals extraction from electronic waste (Cui and Roven 2011). Figure 4 shows the detailed processing steps in detail based on industrial schemes and shows all product streams and subsequent treatments of waste streams.

Another method is vacuum pyrometallurgy, which can recover REEs from the electronic wastes with the difference in pressure using sublimation or distillation. This process is found more useful for the separation of metals such as Bi, Sb, Pb, and other heavy metals through the process of metallurgy as well as recovery of copper from the mixture of molten slats (Flandinet et al. 2012; Zhan and Xu 2009). It also has some limitations for the recovery of the REEs from electronic wastes. Ghodrat et al. (2016) showed that this process has better economic feasibility and can recover metals annually to about 30,000 tones/year. They claimed that the annual recycling capacity of the plant depends upon the internal rate of the return and cost-benefit ratio. However, more research must be carried out in the future for improving the process performance by considering different process configurations, process intensification, and cheaper feedstock (Ghodrat et al. 2016). Table 4 shows some pyrometallurgical methods for the recovery of metals from electronic waste.

Hydrometallurgical

Hydrometallurgy is a chemical method that can be used for the extraction of metals from electronic waste. In this process, REEs, such as gadolinium, yttrium, and lanthanum, are extraction from electronic waste using chloride solution (Baba 1987; Gloe et al. 1990; Macaskie et al. 2007; Ogata and Nakano 2005; Quinet et al. 2005; Wang and Wai 2005; Rodríguez-Rodríguez et al. 2014; Tesfaye et al. 2017a, b; Abdelbasir et al. 2018). Cui and Zhang, 2008a, b) reported that recovery of these elements from electronic waste has some drawbacks due to the generation of sludge, heavy metal pollution, and toxicity. However, Tuncuk et al. (2012) showed that hydrometallurgical processes could achieve high efficacy for the recovery of metals from printed circuit boards. They have also reported the limitation of this method as it can recover only Li and Co metals, not other metals. The extraction and recovery of REE metals from electronic waste using the hydrometallurgy has two steps. The first step is the leaching of metals from electronic waste using mineral acids. For example, Silveira et al. (2015) compared the efficiency of leaching of indium metal from the electronic waste of LCD panel using different acids such as H_2SO_4, HNO_3, and HCl, and the mixture of different acids (like HCl + H_2SO_4 and HCl + HNO_3). They found a higher yield of indium leaching by using HCl and a combination of HCl + HNO_3 than other

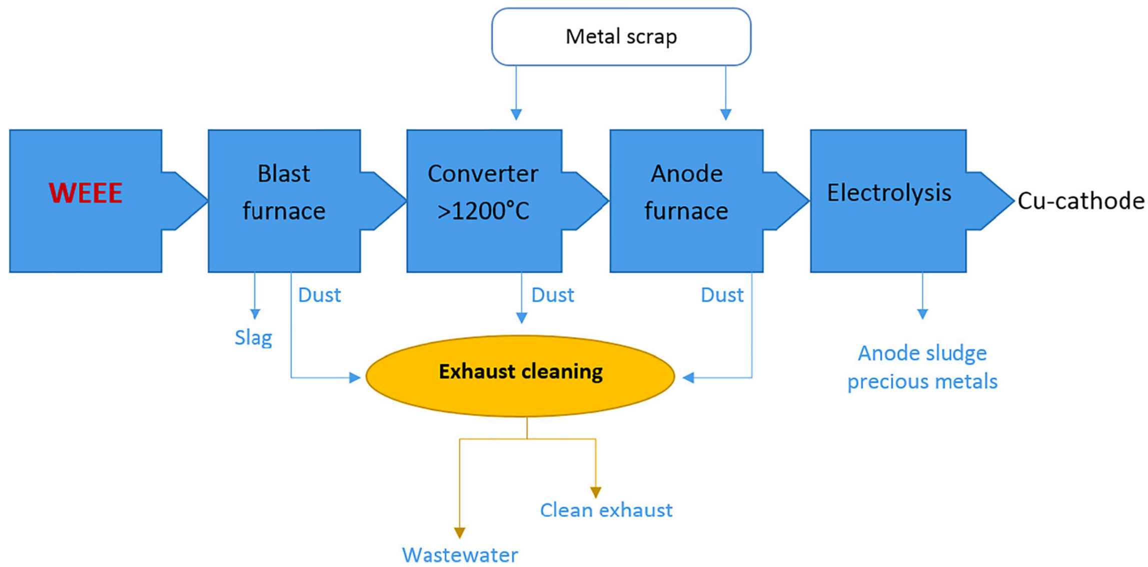


Fig. 4 Pyrometallurgical processing scheme based on industrial copper/WEEE smelting process

mineral acids, probably due to the lower solid to liquid ratio of the HCl (Silveira et al. 2015). This was also supported by Savvilotidou et al. (2015). The leaching of metals from electronic waste can be affected by different parameters such as redox potential, leaching kinetic, lixiviant type, particle size, pH, temperature, and agitation (Savvilotidou et al. 2015).

The second step of the process is the recovery of the dissolved metals from the leachate using adsorption, liquid-liquid extraction, cementation precipitation, and electro-winning. This method is found highly efficient for the recovery of the metals using precipitation and ionic liquid. However, the recovery of metals using precipitation leads to the generation of secondary sludge. Moreover, many studies have shown that recovery of metals using electro-winning and adsorption could solve the issue of sludge production (Anastopoulos et al. 2016; Fujiwara et al. 2007; Huang et al. 2015; Kucuker and Kuchta 2018; Li et al. 2015c). In general, some various methods of hydrometallurgical for the recovery of REEs from WEEE are presented in Fig. 5; Table 5.

Greener physical treatment with supercritical water

In this process, REEs are extracted from electronic waste by using supercritical water and acid leaching (HCl) (Xiu and Zhang 2009). Xiu et al. (2013) showed that supercritical water oxidation and supercritical water depolymerization could recover Cu about 99% and Mn, Sn, Cd, Cr, and Zn about 90%. Similar results were also reported by Liu et al. (2016b) and Xiu et al. (2015), who showed that this method is highly useful for the recovery of precious metal. However, one of the disadvantages of integrating supercritical water with acid leaching for recovery of REEs is the requirement of high temperature (about 420–440 °C). This leads to developing greener

Table 4 Recovery of REEs by Pyrometallurgy from WEEE

Process	Targeted material	Efficiency (mg/g)	Reference
Smelting, copper leaching & electro-winning and platinum group metals refiner	Cu, Au, Ag, Pt, Pd, Se, Te, Ni	High recovery for precious metals and copper	Veldhuizen and Sippel 1994)
Precious metals refining for recovery upgrading in the copper converter and refining	Cu, Ag, Au, Pd, Ni, Se, Zn, Pb	High recovery for precious metals and copper	Lehner 2003)
Copper and precious metals following the copper collector to be recovered to the copper smelter in the zinc fuming process	Copper and precious metals	Almost complete recovery of copper and precious metals	Mark and Lehner 2000)
Smelting operation of WEEE was tested to replace coke as a reducing agent and energy source	Metals in electronic scrap	A high percentage of iron	Cui and Zhang, 2008a, b)

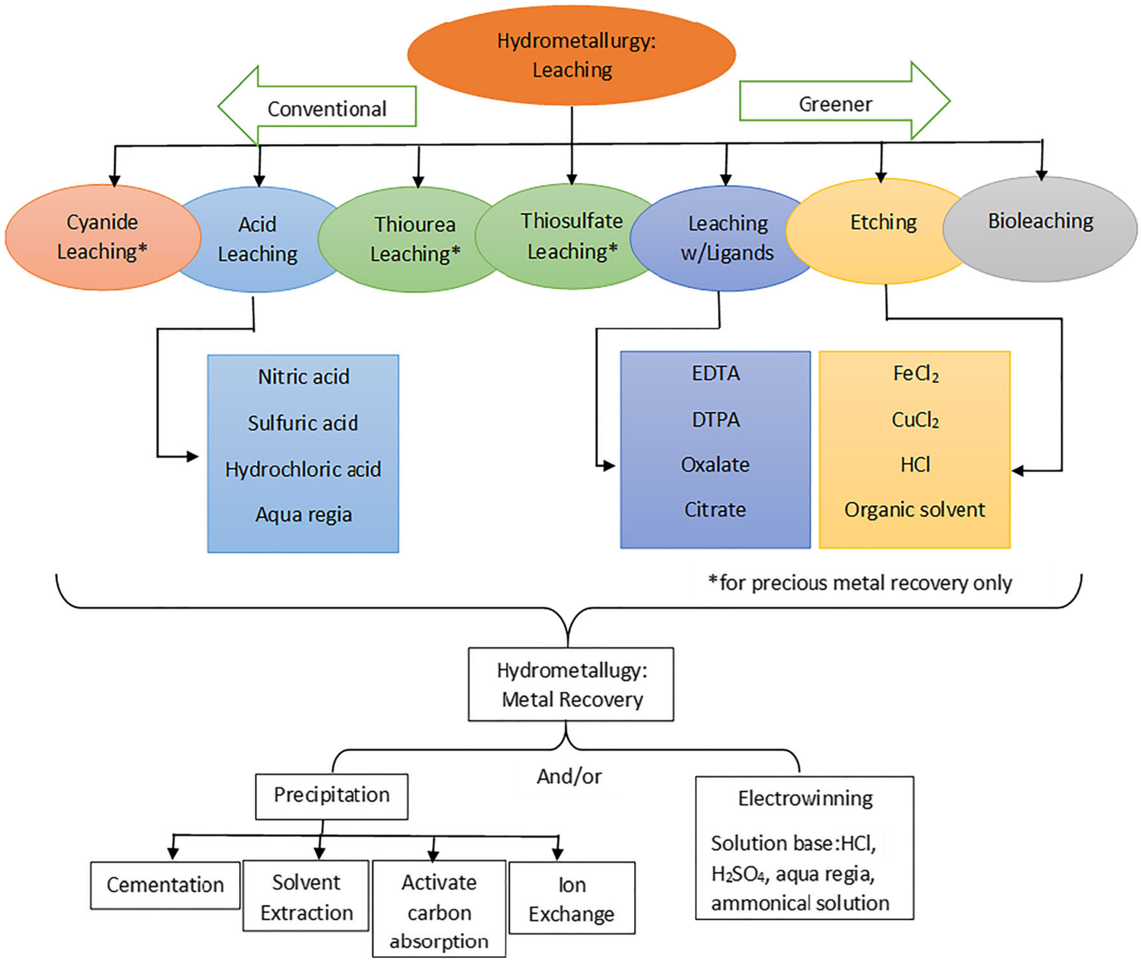


Fig. 5 Summary of hydrometallurgical leaching and metal recovery techniques

treatment processes of supercritical CO_2, having low critical temperature and low toxicity to solve the need. Figure 6 shows the general methodology of a sub- and supercritical water treatment for waste WEEE.

Greener physical treatment with supercritical CO_2

Calgaro et al. (2015) recovered copper metal from electronic waste using supercritical CO_2 integrated with a piranha acid leaching (a mixture of sulfuric acid-water and hydrogen peroxide). The results showed that the supercritical CO_2 extraction process could recover 90% of copper in 20 min, being more effective when compared with atmospheric pressure extraction. Based on the above findings, the recovery of REEs from electronic waste using supercritical CO_2 is very useful. However, a complete understanding of the mechanism of supercritical CO_2 solvent system is still needed, as well as improving its efficiency for recovery of REEs. Extensive research must be carried out in the precise effects of supercritical CO_2 on the extraction of metal, transport phenomena, and leaching kinetics properties. There is a need to integrate a supercritical CO_2 method to other approaches for enhancing the substantiality of this separation process.

Cryo-milling

Cryo-milling is another physical treatment approach, which is a green and novel technology for the recycling of REEs from electronic waste. In this process, electronic waste is degraded into nanoparticle sizes using a ball milling machine, which is operated at low temperatures. This leads to an increase in the efficiency of the separation of oxides, polymers, and meal constituents (Tiwary et al. 2017). Moreover, this approach is an eco-friendly process due to the generation of small volumes of waste and the need for low temperatures, so that it is feasible to apply on a large scale. However, it needs more energy when compared to hydrometallurgical methods. Moreover, further research is still necessary to improve the capacity of the cryo-milling process, by developing a mechanical process

Table 5 Recovery of REEs by Hydrometallurgical from WEEE

Leaching agent	Targeted material	Efficiency	Reference
H_2SO_4-HNO_3-H_2O-NaOH, H_2SO_4-HNO_3-H_2O-NO_X	Cu	> 99.9%	Lekka et al. (2015)
Phosphor powder in HNO_3, HCl, or H_2SO_4	Y and Eu	Over 90%.	Tanvar and Dhawan (2019)
Hydrogen peroxide and sulfuric acid	Co	99%	Nayl et al. (2017)
H_2SO_4	Y	85%	De Michelis et al. (2011)
2 M H_2SO_4	La and Ce	35% La and 35% Ce	Innocenzi and Vegliò (2012)
4 M acid (HNO_3 + H_2SO_4) mixture	Y and Eu	96.4% of Y and 92.8% of Eu	Rabah (2008)
HCl	In	60%	Savvilotidou et al. (2015))
H_2SO_4	Nd	95%	Lee et al. (2013)
H_2SO_4	Nd	99%	Yoon et al. (2014)
H_2SO_4	Y	99%	Innocenzi and Vegliò (2012)
H_2SO_4	In	99%	Li et al. 2011a, 2011b)
HNO_3	Nd and Dy	98% Nd and 81% Dy	Rabatho et al. (2013)
Ammonia and ammonium sulphate	Co	96.3%	Zhang (2005)
Baking followed by H_2O and H_2SO_4 leaching	La, Ce, Nd, Pr, and Sm	80.4% La, 98.8% Ce, 98.2% Nd, 98.5% Pr, 99.2% Sm were leached	Marra et al. (2018)
Ammonia-ammonium salt system containing Cu (I) and Cu (II) ammonia complexion	Cu	100%	Koyama et al. (2006)
HNO_3	Ga	99%	Hu et al. (2015)
Glycine + ascorbic acid	Co	95%	Nayaka et al. (2016)
Succinic acid + H_2O_2	Co	99%	Li et al. 2015a, b)
Tartaric acid + H_2O_2	Co	98%	He et al. (2017)

which is economical and sustainable for the treatment of electronic waste.

Recovery of REEs methodologies to aqueous systems

For recovering REEs from aqueous systems, generally, two approaches (i.e., solid and liquid-liquid extraction) are applied. Hidayah and Abidin (2017) compared the extraction of REEs using solid-liquid extraction over liquid-liquid mining. The results showed that the Liquid-liquid extraction of REEs using aqueous and organic solvent could separate the desired metals by attraction from one side of the liquid phase to the other liquid phase. This process can be applied extensively at a larger scale. In this technology, metals and non-metals are separated using a physical or hydrometallurgical method in which the electronic wastes are dissolving in a liquid phase for the recovery of metals and non-metals (Hidayah and Abidin 2017).

Moreover, it can also recover the metals and non-metals from electronic waste using chemical or pyrometallurgical by the heating process (Kaya 2016). Similar results were also reported for the recovery of metals from electronic waste (Dutta et al. 2016; Hidayah and Abidin 2017). However, this method has some disadvantages, such as loss of precious metals, generation of a large amount of slag that can lead to more challenges in the recovery and extraction of some metals (Kaya 2016; Liu and Jacquier 2006). However, the liquid-liquid extraction approaches are highly applicable on a large scale for the extrication and recovery of metals when compared to the solid-phase extraction. Moreover, these approaches also have some challenges. These challenges are the inability to extract polar compounds, impurities in the final product, emulsions, loss of extractant into the aqueous phase, and low purity of the products. Another challenge is the disposal of toxic or flammable chemicals, which is difficult and time-consuming (Hidayah and Abidin 2017; Fisher and Kara 2016). Figure 7 demonstrates the outlines of the two foremost metal separation and recovery processes, pyrometallurgy, and hydrometallurgy.

Carbon-based nanomaterials applied in solid-phase extraction

Currently, the use of carbon nanostructured materials for the recovery of REMs in solid-phase extraction gets more attention around the globe. Carbon has a variable oxidation number, which leads to the formation of different allotropic forms such as graphene oxide, graphene, graphite, carbon nanofibers, carbon dots, and carbon nanotubes, as shown in Fig. 8.

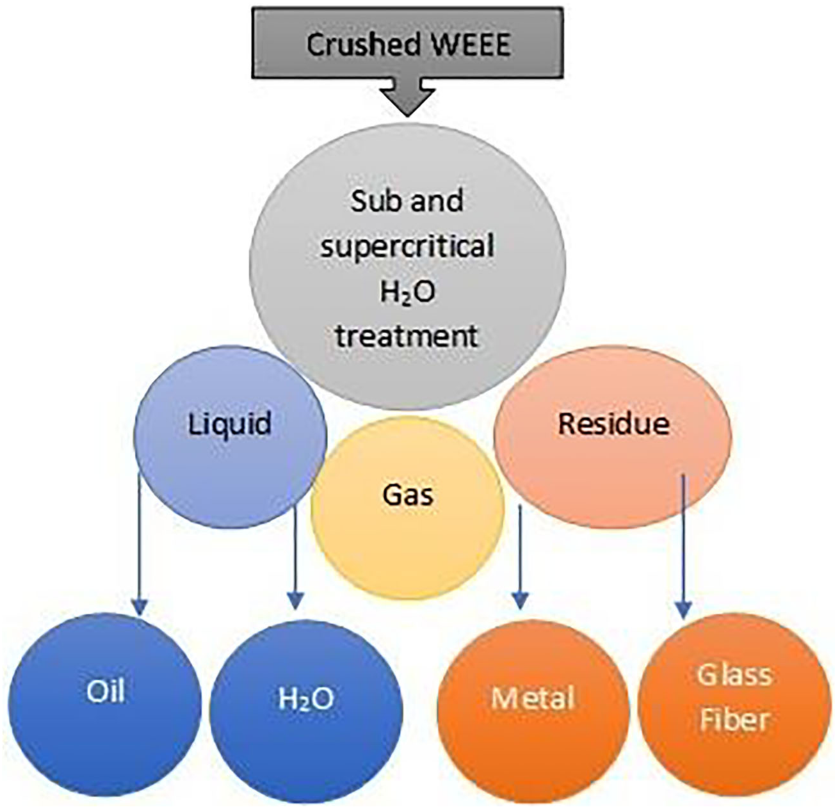


Fig. 6 Sub- and supercritical water treatment approach for waste WEEE

These carbonaceous materials have a high oxidation surface with a more hydrophilic surface. They contain more oxygen functional groups like carboxyl groups and hydroxyl and carbonyl. Therefore, they enhance the recovery of REEs. Their highly complex structure allows further modification of their surface for the sorption of REEs by electrostatic force (Pyrzynska et al. 2016).

Graphene-based composites

Graphene oxide is one of the two-dimensional nanocomposite materials. It has excellent mechanical properties with a large surface area, thermal and electronic conductivity, and hydrophilic properties (Yu et al. 2015). It can be efficiently used for the recovery of REEs from electronic waste. REEs have a high affinity towards the O donors of the graphene oxide (Yu et al. 2015). Consequently, the REEs can adsorb on the O-based surface functional group (Guerrero-Contreras and Caballero-Briones 2015; Yu et al. 2015). Many researchers have reported recovery of the REEs such as europium, cerium, gadolinium, scandium, yttrium, lanthanum, and neodymium from electronic waste using batch and column experiments at room temperature (Chen et al. 2014a, b; Fakhri et al. 2017; Farzin et al. 2017; Su et al. 2014). These studies have shown that the use of nanocomposites is useful for the recovery REEs in the elemental form with no competitive ions because the adoption of REEs is dependent upon the pH (Yao et al. 2016). They also reported effective adsorption of REEs on the surface of graphene oxides. Therefore, more research must be conducted in the future in the optimization of the pH of the solution (Sun et al. 2012; Sun et al. 2013; Li et al. 2014; Li et al. 2015a; Xie et al. 2016; Yao et al. 2016; Ashour et al. 2017; Kilian et al. 2017).

Carbon nanotubes

Carbon nanotubes are one-dimensional nanocomposites and like the other nanotubes. They can also use for the recovery of REE (Behdani et al. 2013; Kilian et al. 2017). Koochaki-Mohammadpour et al. (2014) and Yadav et al. (2015) reported that carbon nanotube composites are effective for the recovery of REEs using distilled water media. They also reported that the sorption of the REEs using this process was dependent upon the time of resident, pH, and temperature. In addition to this, the recovery of REEs is also affected by the type of carbon nanotubes. Chen et al. (2009) and Li et al. (2015b) have found that the recovery of REEs from the electronic waste using oxidized multi-walled carbon nanotubes (O-MWCNTs) is cheaper and more efficient than single-walled carbon nanotubes (SWCNTs-oxidized). They reported that the sorption of REEs using CNTs could increase efficiency by introducing or affording magnetic property such as Fe_3O_4

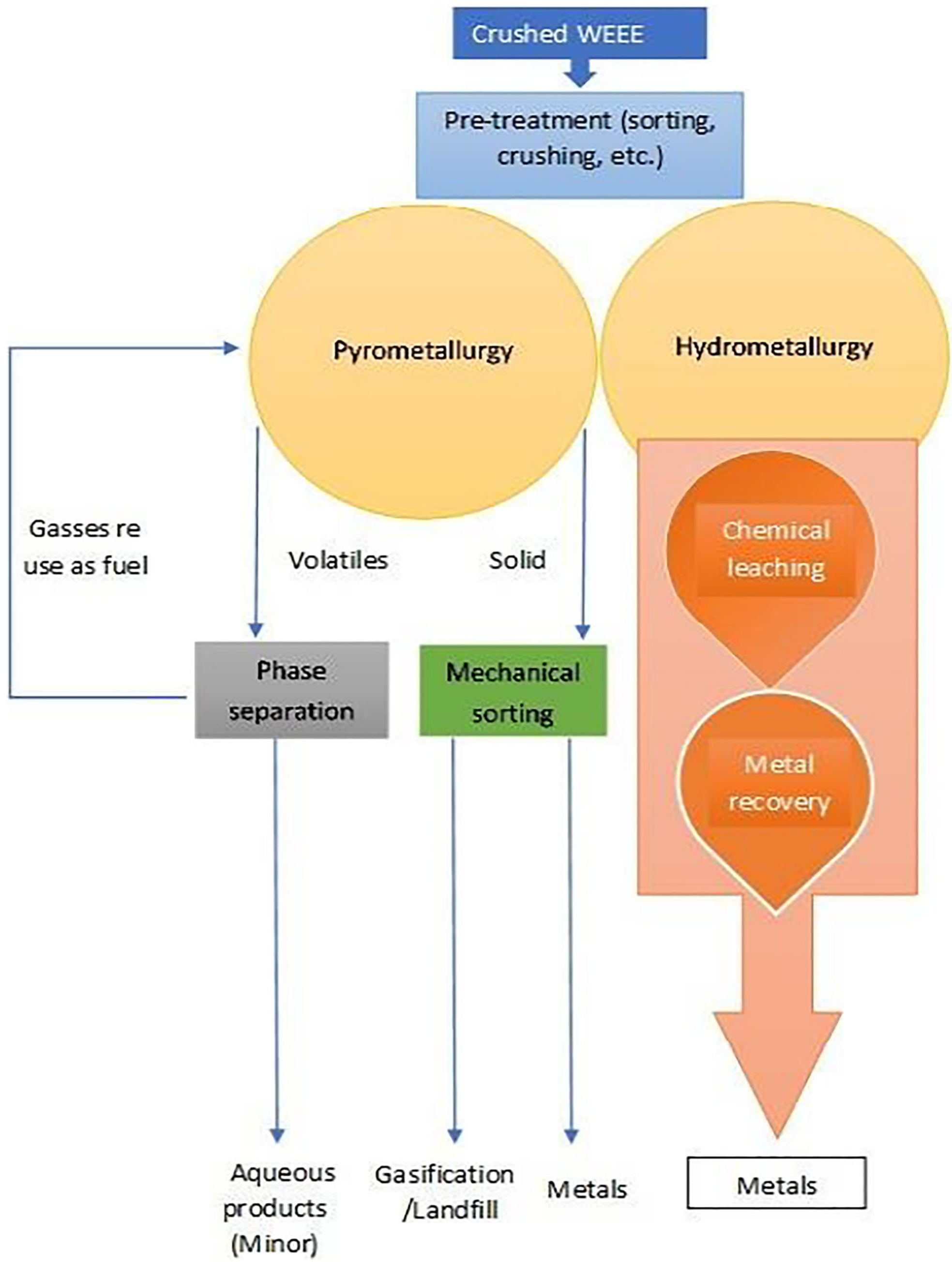


Fig. 7 Summary for the recovery and separation of REEs methodologies to aqueous Systems

or chitosan, due to several functionalizations. Chen et al. (2008), Chen et al. (2009), and Fan et al. (2009) reported that recovery of REEs such as Eu (III) in mono-elemental form was about 600 mg/l and (qm) to about 5000 mg/l from electronic waste.

Other carbon materials

The other carbonaceous materials, i.e., carbon black, mesoporous carbon, fullerenes, carbon nanofibers, and activated carbon, can also be used for REEs recovery from electronic waste. Generally, they do not belong to carbon nanotubes or graphene families (Agrawal 2007; Chen et al. 2007a; Chen et al. 2007b; Pyrzynska et al. 2016). In many studies, other carbon materials are found very effective for the recovery of REEs. However, the recovery depends on the contact time and temperature, as is the case also for other nanocomposites (Gad and Awwad 2007; Sun et al. 2012; Smith et al. 2016; Marwani et al. 2017). These materials have excellent physical and chemical properties such as a large number of oxygen groups and large surface area, as well as magnetic behavior, which can enhance the sorption of metals from aqueous solution.

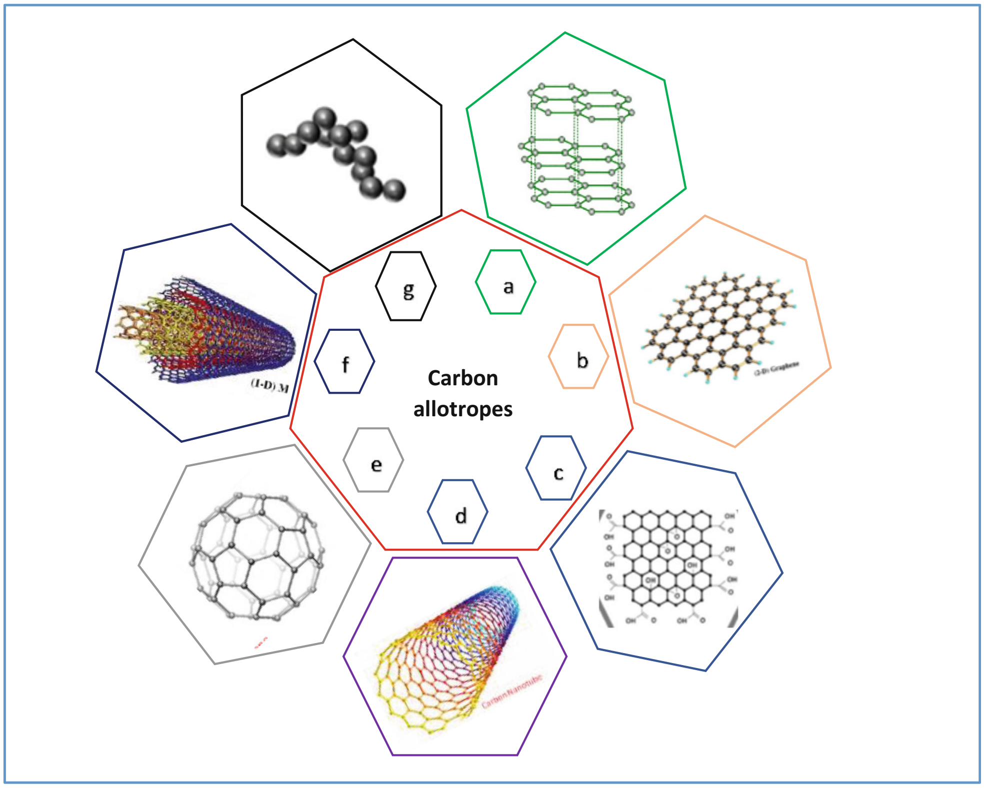


Fig. 8 Allotropic forms carbon: (**a**) graphite, (**b**) graphene, (**c**) graphene oxide, (**d**) carbon nanotube, (**e**) fullerene, (**f**) carbon nanofibers, (**g**) carbon dots

Electrochemical approaches for the recovery of REE from electronic waste

Electrochemical processes (ECP) can be used either for elements extraction or enrichment, within the scope of metal recovery from WEEE. When used as a method for REE extraction from WEEE, scraps may serve as the anode and the application of an electromotive force drives the occurrence of non-spontaneous chemical reactions (Abbasalizadeh et al. 2017; Prakash et al. 2016). In the process, water is oxidized in the anode, producing H^+ (Eq. (1)). Proton accumulation in the anode results in pH decrease, dissolving metals (Vermesan et al. 2020). Electrons flow towards the cathode by an external circuit, and their consumption for water electrolysis takes place in cathodic reactions, generating hydrogen gas and hydroxide ions (Eqs. (2) and (3)) (Prakash et al. 2016). To keep the charge balance, anions and cations migrate between the chambers (Maes et al. 2017). The process results in a solution containing a mix of REE, for further separation.

$$2H_2O \rightarrow 4H^+ + O_2 + 4e^- \quad (1)$$

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \quad (2)$$

$$2H^+ + 2e^- \rightarrow H_2 \quad (3)$$

Complementary, side reactions can occur depending on the system such as the oxidation of iron and/or chloride when they are present (Prakash et al. 2018a). The deposition of non-REEs (iron, nickel, cobalt …) is also probable, according to the type of scrap and cathode composition (Prakash et al. 2016).

ECP may also be applied as a complementary treatment during REE purification. For instance, Prakash et al. (2018a) used ECP to oxidize Fe(II) in the leaching liquor, before REE precipitation. O'Connor et al. (2017) reported an increase in the recovery of REE by increasing voltage up to 3.0 V in an electrochemical filtration system. The use of electrodialysis has also been tested. Anion exchange membranes proved to prevent migration and reduction of Fe(III), reducing co-precipitation of iron ions with REE (Prakash et al. 2018b).

Most results found in the literature related to REE recovery from WEEE via ECP are focused on Nd-Fe-B magnets. Examples are displayed in Table 6. A great advantage of ECP is the low consumption of chemicals in comparison to hydrometallurgical processes (Xu et al. 2020).

Sustainability of REE recovery technologies

The recycling or recovery of REE was reported to present significant economic benefits (Debnath et al. 2018). However, their removal and separation need self-evident chemistry to purify targeted REEs from WEEE. Recently, hydrometallurgical methods were recognized as profitable

Table 6 Application of electrochemical process for rare earth elements (REEs) recovery from electronic waste

Type of WEEE	Sequence of processes	Electrodes composition	Results	Reference
Nd-Fe-B magnets	Electrochemical leaching of Nd-Fe-B scraps and Fe deposition, followed by Nd precipitation with sodium sulphate	Cathode: Cu Anode: Nd-Fe-B	Leaching: 99.9% for 25 mA cm^{-2} Precipitation: Nd (93.7%), Dy (3.1%), and Pr (2.6%), with high purity (99.4%)	Xu et al. (2020)
Nd-Fe-B magnets	Electro-leaching in the presence of H_2SO_4 or H_2SO_4 + citric acid	Cathode: Cu Anode: Nd-Fe-B (inside a 3-d printed Ti basket)	Approximately 0.25 g of Nd, 0.08 g of Gd, 0.6 g of Pr, using 0.1 M H_2SO_4 and 10 A/dm^2, for a leaching time of 60 min	Makarova et al. (2020)
Nd-Fe-B magnets	Acid leaching with HCl, followed by in situ electro-oxidation (Fe(II) to Fe(III)) and precipitation with oxalic acid to recover REE	Cathode: Ni Anode: Ti/Pt Ag/AgCl reference electrode	98% recovery of REE as oxides, with purity higher than 99%	Prakash et al. (2018a)
Nd-Fe-B magnets	Partial leaching with HCl, followed by anion exchange membrane and selective precipitation with oxalic acid.	Cathode: stainless steel wire mesh Anode: mixed metal oxide (35% Ta & 65% Ir) coated titanium electrode Anolyte: partial leachate + undissolved magnet waste Catholyte: sodium chloride solution	Retention after electrolysis at 125 Am^{-2} (%): 95.0 ± 2 (Nd), 95.5 ± 1 (Dy), 98.0 ± 0.5 (Pr), 98.5 ± 0.5 (Co). Purity of REE oxides after precipitation: 99.5%	Prakash et al. (2018b)
Simulated leaching liquor containing Cu and Eu	Multistage electrochemical filtering	Cathodic and anodic carbon nanotube filters	97 ± 0.1% Cu and 65 ± 0.3% Eu recovery	O'Connor et al. (2017)

from an economic point of view for REEs recovery (Cui and Zhang, 2008a, b). These methods do not need extra-energy sources as the energy demand is quite low. Furthermore, these methods allow economizing costs related to the disposal of combustion residue in landfills or treatment, storage, and disposal facilities (Iannicelli-Zubiani et al. 2017). The bio-metallurgical method is recognized as another cost-effective REE processing using bacteria, fungi, or algae to retrieve metals. The cost of bio-metallurgy methods depends on the return from bacterial strain and the cost of chemicals, the medium needed for promoting the culture, and the culture conditioning, though further studies are needed to scale-up and for process intensification to turn it into sustainable technology. Adding recycled REEs as a new source to the supply chain is anticipated to overcome environmental pollution and energy costs linked with their primary mining and separations.

Conclusions and perspectives

This review paper highlights the future perspective for the recovery of REEs from WEEE wastes, which is increasing at an alarming rate all over the world. The recent development of various novel technologies for sustainable and effective recovery of REEs from electronic wastes is getting attention among policymakers and scientists. This paper also highlights the research progress on different efficient processes for the recovery of metals from electronic wastes through bioleaching, biosorption, siderophores, hydrometallurgy, pyrometallurgy, and carbon-based material. In addition to this, if these technologies are used properly, they can help not only in achieving sustainable development goals but also balance between social, economic, and environmental sustainability.

Hydro- and pyrometallurgy are the widest applied technologies for WEEE secondary treatment. Despite their known effectiveness, issues as energy and chemical consumption, as well as liquid and/or gaseous emissions, should be addressed. Greener and promising alternatives are bio-based processes. The use of microorganisms for bioleaching may lead to the high recovery of REEs. However, slow kinetics and low solid-liquid ratio still hinder large-scale applications. Siderophores, in turn, are a green approach that seems to be very promising for the recovery of REEs present in small quantities in WEEE. Electrochemical processes may also be an alternative to reduce the consumption of chemicals and wastewater generation. Especially when associated with other methods, they have shown to result in high purity rare earth. Carbon-based materials are also an alternative that may lead to increase products' purity.

Electronic waste is one of the emerging problems in developed and developing nations worldwide. If they are not adequately treated or recovered and stored, they can lead to damage to the environment, particularly to the aquatic environment. Moreover, they have high economic value and high content of the REEs. So, they must be recycled. Currently, the recovery of REEs is not sustainable. The development of appropriate and environmentally friendly novel technology to recover REEs and treat electronic wastes are urgently required. Future research must be emphasized on the needs of automated and green processes. The technology options for the recovery of materials from WEEE must have an impact on the macro- and micro-perspectives of process feasibility and economic gain with sustainability.

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