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To cite this version:

Arda Işildar, Eric D. van Hullebusch, Markus Lenz, Gijs Du Laing, Alessandra Marra, et al.. Biotechnological strategies for the recovery of valuable and critical raw materials from waste electrical and electronic equipment (WEEE) – A review. Journal of Hazardous Materials, 2018, 362, pp.467-481. 10.1016/j.jhazmat.2018.08.050 . insu-02549280

HAL Id: insu-02549280

https://insu.hal.science/insu-02549280v1

Submitted on 6 Aug 2020

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Journal of Hazardous Materials 362 (2019) 467–481

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Biotechnological strategies for the recovery of valuable and critical raw materials from waste electrical and electronic equipment (WEEE) – A review

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ARTICLE INFO

Keywords:
Biotechnologies
Bioleaching
Biosorption
Bioprecipitation
Critical metals
Electronic waste

ABSTRACT

Critical raw materials (CRMs) are essential in the development of novel high-tech applications. They are essential in sustainable materials and green technologies, including renewable energy, emissionfree electric vehicles and energy-efficient lighting. However, the sustainable supply of CRMs is a major concern. Recycling end-of-life devices is an integral element of the CRMs supply policy of many countries. Waste electrical and electronic equipment (WEEE) is an important secondary source of CRMs. Currently, pyrometallurgical processes are used to recycle metals from WEEE. These processes are deemed imperfect, energy-intensive and non-selective towards CRMs. Biotechnologies are a promising alternative to the current industrial best available technologies (BAT). In this review, we present the current frontiers in CRMs recovery from WEEE using biotechnology, the biochemical fundamentals of these bio-based technologies and discuss recent research and development (R&D) activities. These technologies encompass biologically induced leaching (bioleaching) from various matrices,biomass-induced sorption (biosorption), and bioelectrochemical systems (BES).

1. Introduction

Electronic waste, E-waste or waste electrical and electronic equipment (WEEE) refers to discarded devices that are at the end of their economic use and cannot be utilized by consumers anymore. The product spectrum of electrical and electronic equipment (EEE) expanded rapidly, coupled to an increase in consumer demand for electronics and affordability. This resulted in an unprecedented global WEEE generation. WEEE constitutes the largest and fastest growing fraction of municipal waste [1], and reached a global total of 41.8 million tons per annum in 2014 [2]. The EU-28 plus Norway, Iceland, Liechtenstein, Switzerland, and Turkey are the largest WEEE generators with a total of 9.8 M tons and 20.4 kg/person/year in average, along with the United States (7.1 M tons and 22.3 kg/person) and China (6 M tons, 4.4 kg/person) [2,3]. WEEE generation is directly correlated with gross domestic product (GDP) [4], and the per capita generation increased particularly in the developing countries [5].

Improper management of WEEE is an alarming global environmental problem due to the presence of a large variety of toxic substances embedded in the devices [6]. Despite preventive legislation, most WEEE is still poorly managed, either landfilled, or transferred to developing countries either through legal or unregistered routes [7]. In Europe approximately 35% of WEEE is recycled, and the rest is landfilled, exported or lost [4,8]. According to the current EU directive regarding WEEE [203], approximately 45% of WEEE should be collected in 2016, and the minimum collection rate annually shall be 65% in 2019 (European Commission, 2012). Nevertheless, collection rate in EU-28 countries are higher than those of other high GDP countries.

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https://doi.org/10.1016/j.jhazmat.2018.08.050
Received 17 January 2018; Received in revised form 14 August 2018; Accepted 16 August 2018
Available online 17 August 2018
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Whereas transboundary movement of WEEE from developed to developing countries had become the norm in 2000s, the situation is now changing due to an increasing public interest under the umbrella of the Basel Convention and the regional precautionary measures taken by the governments.

WEEE is an important secondary source of valuable and critical metals. Perpetual innovation of consumer EEE resulted in highly variable material properties and shape of the products, with an increasing complexity [9]. Thus, the elemental composition of the discarded devices is also highly variable and complex [10]. A modern smartphone includes up to 58 elements at various concentrations and chemical composition [11]. Thus, novel recycling strategies should consider selectivity for metal recovery from these complex materials. The proper implementation of novel resource recovery-oriented recycling strategies may contribute to controlling the environmental risks associated with improperly managed WEEE. In the recent years, considerable research efforts have been carried out to develop environmentally friendly biotechnological processes. Selectivity towards individual metals, cost-effectiveness and eco-innovation are the potential advantages of biotechnological processes [12]. They are foreseen to play a considerate role in sustainable development, particularly for the metallurgical, chemical and waste processing sectors [13].

Currently, high-grade WEEE is treated in high temperature pyrometallurgical facilities, to recover the valuable metallic fraction of the end-of-life devices (Ebin and Isik [196]). Several investigations on hydrometallurgical metal recovery from WEEE have also proven successful and financially feasible at various technology readiness levels [14,15]. Biotechnologies may offer promising alternatives to pyrometallurgical technology in metal recovery from post-consumer waste. Biohydrometallurgy is already an established route to process primary ores of many metals [16,17] and may play an important role in the urban mining of critical raw materials in the future. Selectivity towards critical and valuable metals may be a major advantage of biotechnologies over conventional chemical recovery methods [18]. Further, they may offer advantages in cost-effectiveness and lower environmental impact [19]. This review presents the latest developments in global WEEE generation and critical metals contained therein and focusses on the use of biotechnologies to recover both critical and conventional metals from these waste streams. Specifically, this review focusses on recent developments in bioprocessing by such diverse biotechnological strategies as autotrophic and heterotrophic bioleaching, biosorption, bioprecipitation and bioelectrochemical recovery.

2. Global WEEE management

2.1. WEEE classification, hazards, and global generation

WEEE encompasses a wide range of discarded devices, and is classified per product type and legislative relevancy. WEEE is grouped into 10 primary categories according to the WEEE Directive by the European Commission (2012/19/EU), i.e. (i) large household appliances, (ii) small household appliances, (iii) information technology and communication (ITC) equipment, (iv) consumer electronics, (v) lighting, (vi) electrical and electronic tools, (vii) toys, (viii) leisure and sports equipment, (ix) medical devices, and (x) automatic dispensers. These collection categories also exist in actual WEEE collection and management practice [20]. These ten major product categories are further grouped into 58 sub-categories, representing approximately 920 products.

Due to the nature of minerals, energy and chemicals used for production, the energy usage during use, and materials landfilled at the end of life, WEEE is a rapidly growing global environmental problem. A considerate share of WEEE generated is landfilled or shipped to developing countries where it possesses a significant hazard to the environment and local communities [21]. The hazards are associated with the presence of heavy metals, brominated flame retardants (BFRs),

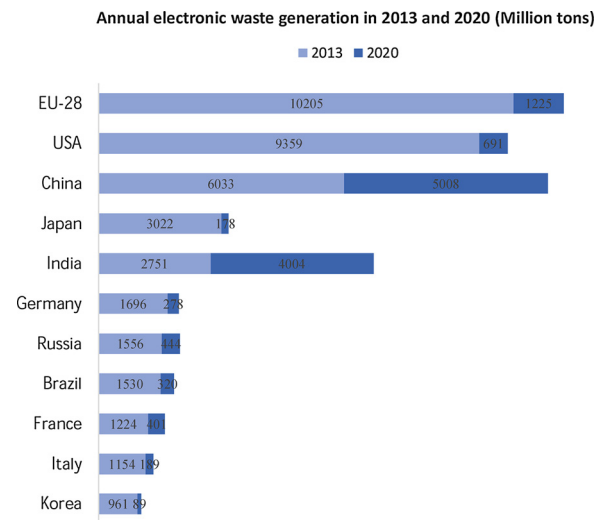

Fig. 1. Annual electronic waste generation in 2013 and future prediction in 2020.

polybrominated diphenyl ethers (PBDEs), dioxins and other potentially harmful substances either contained in or formed during waste processing [10]. In addition, about 50% of personal computer components contain hazardous arsenic (As), hexavalent chromium (Cr(VI)) and mercury (Hg) [3].

The quantification of WEEE volumes is a prerequisite for the development of sustainable solutions. Challenges include the lack of reliable data, sensitivity-related issues, the dynamic nature of the flows and their constituents (Schluep et al. [187]). This task is particularly unwieldy in developing countries as informal waste management systems are poorly documented and data quality is an issue [22,23]. In addition, lack of quantitative understanding of the amounts involved in transboundary WEEE movement is prevalent [24]. An overview of the total WEEE generation in 2013 and its forecast for 2020 is shown in Fig. 1 [19,24–26]. Generation of WEEE has exponentially increased due to rapid technological innovations in the electronics sector, coupled with demand growth for electronics in developing countries. In addition, decreasing economic lifespan of electronic devices [27,28] lack of international legislative consensus on WEEE management [29] and affordability played a major role in increasing the generation of WEEE. Lifespan of the electronic decreased to an average of 10 to 2 years and 24 to 9 months for large EEE and mobile phones, respectively [27,30–32]. These issues, coupled with ever-increasing spectrum of devices make WEEE the fastest growing post-consumer waste stream. In 2014, 41.8 million tons of WEEE was generated, and the value is expected to increase to over 50 million tons in 2018 [2]. WEEE occupies an increasing fraction of municipal waste, up to 8% of total municipal waste in developed economies, with saturated EEE markets [33]. Reportedly, a substantial increase in WEEE generation is also expected in developing countries, as a result of the forecasted economic growth [34]. Consequently, regions with large populations and rapid economic growth are expected to become large WEEE producers in the coming years.

Discarded devices are particularly concentrated in urban areas where population density is very high [35]. This leads to an emerging field of research termed as urban mining, in which many waste materials are re-used as a secondary source of materials [36]. Analogous to primary ores, urban mines are distinct with regard to their metal composition and content [32]. Usually, in WEEE derived from “urban mines” metals are found in complex alloys and in their metallic elemental form [37]. This requires a novel approach to sustainably and selectively recover metals from WEEE. Conventional mechanical, pyrometallurgical, hydrometallurgical and bio-hydrometallurgical

processes, and often a combination of these are proposed for metal recovery from WEEE [38–40]. Biotechnologies might find a niche in this area with their lower waste production, emissions, and carbon footprint.

2.2. Critical metal content of WEEE

Primary sources of critical metals, such as rare earth elements (REE) and platinum group metals (PGM), are neither ubiquitously available nor equally distributed in the world. For instance, China produces 87%, 95%, and 95% of Sb, heavy REE (HREE) and light REE (LREE), respectively. Brazil supplies 90% of niobium (Nb), and the USA 90% of beryllium (Be) [41]. Chinese export restrictions on REE in 2009 triggered a raw materials supply issue, and gave momentum to several national and international initiatives (European Commission [204]; [42]; USNRC [205]). The drivers of resource scarcity are associated with (1) the demand of a growing world population striving for a high standard of living, and (2) geopolitical scenarios such as, export restrictions and/or political instability of major producing and reserve-hosting nations, rather than exhaustion of the primary ores. The European Commission (EC) ranked the raw materials in terms of their criticality according to the risks of supply shortage and their economic importance for the first time in 2011 and updates this list regularly [41]. The list currently includes 27 CRM and a number of them are essential for the EEE, in particular emerging green technologies. EEE contain many critical metals including the light and the heavy rare earth elements (LREE, HREE), cobalt (Co), antimony (Sb), tungsten (W), gallium (Ga), germanium (Ge), indium (In), tantalum (Ta), and platinum group metals (PGM), and near-critical elements such as tin (Sn), chromium (Cr), lithium (Li), and silver (Ag). They are essential components of EEE and have an increasing importance in the transition to a green, low-carbon economy. Examples of critical elements and their abundance in respective WEEE units is given in Table 1.

Critical materials (such as Co, HREE, In, Li, LREE, Ni, PGM, and Sb) are essential to the functionality of EEE and can often not be replaced. In turn, printed circuit boards, permanent magnets, lithium-ion, nickel metal hybrid (NiMH) and nickel cadmium (NiCd) batteries, lamp phosphors, liquid crystal displays (LCDs), light emitting diodes (LED), and hard disc drives (HDD) are thus important secondary sources of critical metals (Binnemans et al. [188]; Ueberschaar and Rotter [175],18]). The risk for insufficient supply and scarcity of raw materials is now perceived by many leading companies from different manufacturing industries, particularly, renewable energy, electric vehicles, and consumer electronics. This underlines the importance of developing novel (bio)technologies for recovery of critical raw materials from WEEE, which has been identified as a strategic research direction in the EU.

3. Bioprocessing of WEEE for metal recovery

Biotechnology is an established route for extraction of Au, Ag, As, Co, Cu, Mn, Mo, Ni, U, V, and W, Zn from primary ores (Morin et al., 2006, [17]). Around 15% of copper (Cu) and 5% of gold (Au), and lower amounts of nickel (Ni) and zinc (Zn), are produced using microorganisms (Johnson [206] [12]). Bioleaching was proven to be an applicable technology for processing of primary, in particular the low grade ores. There is an increasing academic and commercial interest in bioprocessing of waste for metal recovery, which can be attributed to its **(1)** potentially better environmental profile, **(2)** ease and practicality of operation, **(3)** better cost-effectiveness, and **(4)** potential for future development. Moreover bio-based technologies could be more selective towards metals, which gives them an additional advantage. The biochemical mechanisms involved in biomining of primary minerals are well understood and explained in detail e.g. by Mahmoud et al. [12]. Brierley and Brierley [45] and Rohwerder et al. [46]. Waste materials, i.e. post-consumer anthropogenic discarded materials, on the other hand, show dissimilarities to primary ores, as most metals are found in their zero valence elemental state in WEEE, often alloyed with other metals [11,32].

3.1. Bioleaching of critical metals from WEEE

Bioleaching of metals is carried out by a largely diverse group of microorganisms, mainly including three groups of microorganisms, namely **(i)** chemolithotrophic prokaryotes, **(ii)** heterotrophic bacteria and **(iii)** fungi [47]. In nature, a large variety of chemolithotrophic and organotrophic microorganisms are involved in bioleaching of ores [48] (Panda et al. [189]). Current state-of-the-art research on metal recovery from WEEE via biotechnology involves both autotrophic (i.e. sulfur-and iron-oxidizers) [49,50] and heterotrophic (e.g. cyanide-producing telluric microorganisms) [51,52]. The fundamentally different chemistry of metals contained in primary (sulfidic) ores in contrast to WEEE implies that different leaching mechanisms underlie. Fig. 2 gives an overview of conventional autotrophic bioleaching of primary ores and heterotrophic and autotrophic bioleaching of secondary raw materials.

3.1.1. Chemolithotrophic autotrophic bioleaching

Chemolithotrophic organisms utilize atmospheric carbon dioxide (CO_2) as carbon source, and inorganic compounds such as ferrous iron (Fe^{2+}), elemental sulfur (S°) and/or reduced sulfur compounds as energy source [53,54]. In biomining, they are the most widely studied group of microorganisms, and the subject of a considerable amount of fundamental and applied research [55–57]. Most chemolithoautotrophs have a high tolerance for heavy metals toxicity [58], which makes them the most widely used group of microorganisms to process also other

Table 1
Examples of critical metal content of WEEE components.

WEEE component	Metals[a,b] (%, *w/w*)	Concentration (ppm)	Number. of critical and valuable metals	References
Main boards				
Printed circuit boards	**Au**, **Pd**, **Ge**, **Ga**	1 – 100	4	Ghosh et al. [39]; Hadi et al. [10]
	Fe, Al, Ag, Ni, Zn	100 – 10,000		
	Cu	> 10,000		
Batteries				
Li-Ion batteries	**Co, Li**	1 – 100	2	Lee and Pandey [43], Bigum [213], Kim et al. [44]
NiMH batteries	**Co**, **La**, Ni	100 - 10,000	2	
Memory drives				
HDD magnets	**Nd, Pr, Dy**	> 10,000	3	[175], Cucchiella et al. [183]
Solid state drives (SDD)	Cu, Ag, **Au**, Pd	1 – 100	4	
Displays				
Liquid crystal displays (LCD)	**Y**, **In,** Sr	1 – 100	2	Zhang et al. (2015); Cucchiella et al. [183]
Light emitting diodes (LED)	**Au**, Ag, **In**, **Sn**	1 – 100	3	

[a] Only metals with concentration over 1 ppm are given.
[b] Critical metals are highlighted in bold letters.

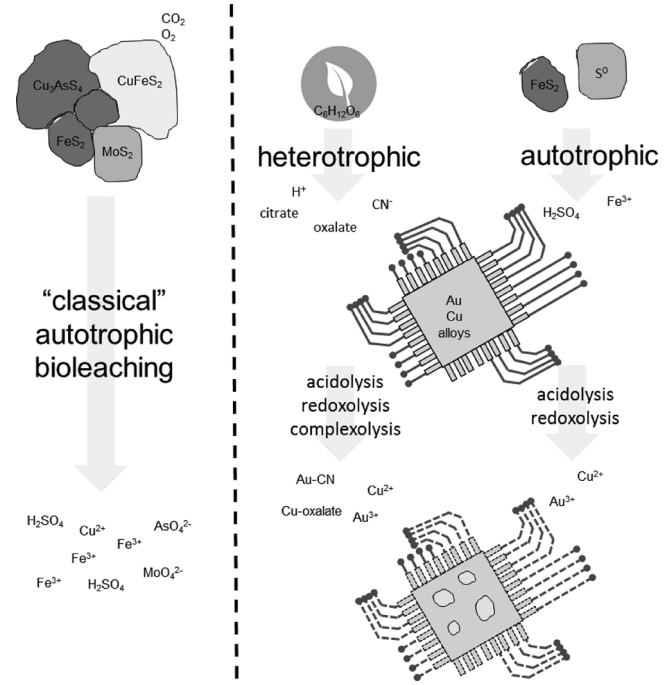

Fig. 2. Conventional autotrophic bioleaching of primary ores and heterotrophic and autotrophic bioleaching of secondary raw materials.

polymetallic sources such as WEEE (after addition of reduced Fe/S minerals, see Fig. 2). *Acidithiobacillus ferrooxidans, Acidithiobacillus thiooxidans* and *Leptospirillum ferrooxidans* are the most extensively studied mesophilic microorganisms in bioleaching communities [59]. On the other hand, thermophilic processes involve microorganisms such as *Acidianus brierleyi*, *Sulfobacillus thermosulfidooxidans* and *Metallosphaera sedula* [60]. These acidophiles thrive on iron-and sulfur-containing ores such as pyrite, pentlandite [$(Fe,Ni)_9S_8$] and chalcopyrite ($CuFeS_2$) at temperatures between 45 and 75 °C.

3.1.2. Autotrophic bioleaching of secondary sources

Autotrophic bioleaching of secondary sources may be misleading from the point that autotrophs cannot grow directly on oxidation / dissolution of the WEEE matrix. However, when mixed to the WEEE substrate, sulfidic minerals such a pyrite [61] can provide energy for autotrophic growth. As in "conventional" autotrophic leaching, microbial oxidation of sulfidic minerals will then result in production of acidity and ferric ions, which can in turn solubilize metals from WEEE. Whereas autotrophic bioleaching of sulfidic ores will ultimately lead to a dissolution of (most of) the matrix, the non-metallic fraction of WEEE will not be dissolved, which is an important difference for processing schemes (Fig. 2). The application of biomining for REE recovery from other secondary sources faces similar challenges due to the particular matrix in which the REEs are embedded. REEs are typically extracted as phosphates (monazite and xenotime) or carbonates (bastnäsite) in the primary ores exploited.

Early studies from between 1980 and 1990 revealed the ability of *A. ferrooxidans* and *Acetobacter* strains in mobilizing REEs from minerals; however, the mechanism of interaction of microbes with rare earths is still not well-known [62]. A limited number of studies on REE bioleaching is currently available; these are mainly focused on the extraction of rare earths from native minerals [63–66]. The only record of a REE bio-heap leaching project is carried out by DNI Metals in Alberta Canada. Economically viable quantities of Sc, i.e. ~5 g/ton was explored in polymetallic sulfides [18].

Research work on chemolithotrophic autotrophic mesophilic bioleaching of metals from WEEE using acids produced by iron- and sulfur-oxidizers is limited. It focused mainly on recovery of transition metals [50,67] and also REEs to a smaller extent [68]. Investigations on autotrophic bioprocessing of WEEE were carried out using moderate thermophiles [69], although recent reports have shown the feasibility of bioprocessing WEEE at ambient temperatures [70,71].

3.1.3. Heterotrophic bioleaching

The development of several biotechnological systems for metal recovery from secondary raw materials, such as REE-bearing waste as red mud, slags, coal ashes and fluorescent powders, was recently initiated ([72]; Potysz et al. [190] [73,74]). As REE-containing waste does not contain metal sulfides, bioleaching via heterotrophic microorganism appears as a promising approach [74]. Moreover, heterotrophic microorganism can tolerate high pH conditions as well as complexed metals in solution [75].

Bacteria, archaea and fungi are typically involved in heterotrophic bioleaching of metals [53]. Compared to acidophiles, heterotrophs tolerate a wider range of pH and are employed for treating moderately alkaline wastes [76]. Research on heterotrophic bioleaching of critical metals from WEEE has been focused on cyanide- and organic acid-generating microorganisms. Cyanogenic bioleaching targets precious metals and the platinum group metals (PGM), i.e. Au, Ag, Pt, Pd, Rh, and Ru which are often not leachable by mineral acids. Critical metals, such as Co, Ga, Ge, Li, Sb, and W, are typically leached from secondary sources using chelation. Heterotrophic bacteria and fungi contribute to bioleaching through biosynthesis of organic acids solubilizing metals (Bosecker [207]; Gadd, 2000). Organic acids; namely acetic acid, lactic acid, formic acid, oxalic acid, citric acid, succinic acid, and gluconic acid can mediate complexolysis (Brandl [209]). To the best of our knowledge, no industrial heterotrophic bioleaching project has been implemented yet. Due to the need for a high supply of carbon and energy that are needed for the metabolic activities of the heterotrophic microorganisms, full-scale application has been implemented yet [18].

3.1.3.1. Heterotrophic bacterial bioleaching of metals. Several *Pseudomonas* strains such as *P. aeruginosa*, *P fluorescens*, and *P. putida*, are involved in bioleaching of valuable metals. They are microbes that are found ubiquitously, typically in soils, and solubilize metals owing to various metabolic products. Biogenically produced cyanide is excreted during growth limitation phase, and provides the cyanide-tolerant microbe a selective advantage [77]. Cyanide excretion occurs in soils with top layers rich in organic matter, where a symbiotic relationship between the plants and the cyanogenic microorganisms occurs [78]. Bioleaching of Cu, Au, Ag, Pt, and Zn by various *Pseudomonas* species from primary ores [79,80], metallurgical slags (Cheng et al. [191]; Potysz et al. [190]), and crushed WEEE [81] has been reported. Complexolysis reactions with cyanide leading to gold solubilization are not only the basis for bioleaching [71,82], but also for conventional gold mining (Akcil et al. [192]).

Chi et al. [83] investigated bioleaching of Au with *Chromobacterium violaceum* from high-grade cell phone PCB. He reached 10.8% (0.46 ppm) Au removal in 8 days with an increase in pH. Ruan et al. [52] identified a new *Pseudomonas* species from a mining region based on 16S rDNA analysis. The new species *Pseudomonas chlororaphis* (PC) removed 8%, 12%, and 52% of the Au, Ag, and Cu from discarded PCB at the optimal conditions of pH at 7, room temperature (25 °C), in 72 h, with the addition of 4.4 g/L glycine as precursor and 2 g/L methionine as catalyzer.

Cu is predominantly found in discarded PCB, and also other types of electronic waste. It competes with the other metals, i.e. Au and Ag, for biorecovery of precious metals, due to preferential complexation and leaching. Işıldar et al. [71] developed and applied a two-step autotrophic/heterotrophic biorecovery strategy to selectively remove Cu and Au. Cu was bioleached by a mixture of autotrophic iron- and sulfur oxidizer *Acidithiobacillus ferrivorans* and *Acidithiobacillus thiooxidans* in the first step with 98.4% removal. In the second step, Au was removed by *Pseudomonas putida* with 44.0% removal at ambient temperature

(25 °C). The relatively low gold removal was related to low cyanide generation of the *Pseudomonas* cultures and a cyanide concentration of 21.4 mg/L did not allow complete gold removal from the waste material.

Several strategies were elaborated to increase the biogenic cyanide production, including sequential nutrient addition [51], medium modification [84], and genetic modification [85]. Ting and Pham [86] investigated the adaptation of cyanogenic bacteria to pH values above 9.5 so as to promote the bioleaching efficacy. *Chromobacterium violaceum*, the most widely studied cyanogenic heterotroph was found to adapt to pH values up to 9.5 [87]. Adapted cells of the cyanogenic microorganism bioleached 18%, 22.5% and 19% of Au at pH 9, 9.5 and 10, respectively, while non-adapted bacteria bioleached only 11% at pH 7. Natarajan and Ting [84] further investigated Au bioleaching efficacy of genetically modified strains, showing highest biogenic cyanide activity (bioleaching of 30% of Au).

Heterotrophic bioleaching of REE from waste materials by biogenic gluconic acid has recently been demonstrated. Three microbial isolates, two bacterial strains identified as *Acinetobacter* and *Pseudomonas* species and a fungal one related to *Penicillum* and *Talaromyces*, were selected along with the industrially known bacterium of *Gluconobacter oxydans* for leaching phosphor powders and spent fluid catalytic cracking catalysts. For phosphor powders, the REE leaching efficiency reached a maximum of 2%, while 49% of total REE was leached out from the fluid catalytic cracking catalyst using cell-free culture supernatants of *G. oxydans* [74]. Heterotrophic bacterial leaching of REE from primary ores and thorium-uranium concentrates (monatize minerals) was investigated. *Pseudomonas aeruginosa* exhibited REE leaching efficiencies of up to 63.5% under optimum conditions [88] while *Acetobacter aceti* showed low extraction efficiency (0.13%) [89], respectively.

Marra et al. [90] investigated the recovery of base metals, precious metals and rare earth elements from WEEE dusts. In the first step, base metals were almost completely leached from the dust in 8 days by *Acidithiobacillus thiooxidans* at acidic conditions. During this step, cerium, europium and neodymium were recovered at high percentages (> 99%), along with La and Y with a yield of 80%. In the second step, cyanide-producing *Pseudomonas putida* recovered 48% of Au within 3 h from by *A. thiooxidans*.

3.1.3.2. Fungal bioleaching. Fungal bioleaching mechanisms involve leaching of metals by organically excreted acids (acidolysis and complexolysis) and change in the oxidation potential of the medium (redoxolysis), or a combination of the three [91–93]. In contrast to acidophilic bacterial leaching, fungal redoxolysis bioleaching takes place at a relatively higher pH, i.e. near-neural or alkaline values (Xu and Ting [197]). *Aspergillus niger* and *Penicillium simplicissimum* are among the most studies microbes in fungal bioleaching of metals from waste material [43]. In a first attempt to mobilize metals from electronic waste Brandl et al. [94] used the latter species to extract Cu and Sn with 65% efficiency, and Al, Ni, Pb, and Zn by more than 95%. After a prolonged adaptation time of 6 weeks, the microorganisms were able to adapt to higher pulp densities of up to 10% (*w/v*). The authors recommended a separate process where the cells are grown in absence of waste material due it is inhibitory effect on growth. Bioleaching of Al, Cd, Cu, Co, Fe, Li, Mn, Ni, Pb, Zn from incinerator ash [95,96] by *Aspergillus niger* and Cd, Co, Mn, Ni, Zn from spent Zn-Mn or Ni-Cd batteries by six *Aspergillus* species [44] were investigated. The authors noted the difference in metal removal efficiency when different carbon sources were used, which is related to the conversion of the given carbon source and the excreted organic acid.

Fungal bioleaching of the critical REE by *Aspergillus ficuum* from primary sources has been investigated as well by Hassanien et al. [88]. In this study, under optimum conditions, 75.4% and 63.8% of REEs were directly bioleached from monazite and a thorium-uranium concentrate, respectively. *Aspergillus ficuum* was subject to another study by Desouky et al. [64], who removed 20% of lanthanum, 33% of cerium and 2.5% of yttrium at pH 3.0 in 24 h by this fungal strain from thorium-uranium concentrates. Brisson et al. [63] used the fungal strains of *Aspergillus niger* ATCC 1015, *Aspergillus terreus* ML3-1 and *Paecilomyces* spp for solubilizing REE using monazite as a phosphate source. Comparison with abiotic leaching experiments indicated the beneficial effect of microorganism presence.

Fungal bioleaching of REE was investigated by Qu and Lian [73], who focused their work on waste red mud material, i.e. the main by-product in bauxite processing for alumina production. Leaching experiments were performed using *Penicillium tricolor* under three different pulp densities (2%, 5%, 10%) and three different bioleaching processes: (1) one step bioleaching, involving microorganism cultivation in presence of red mud; (2) two step bioleaching with a fungal pre-growth in absence of red mud followed by its addition; (3) cell-free spent medium. The maximum leaching extraction of REEs was achieved by the one step leaching method at 2% pulp density, whereas a two-step leaching process exhibited the highest efficiency at 10% pulp density.

3.1.4. Biochemical mechanisms of bioleaching reactions

The exact mechanism of biological metal extraction from waste material has been long debated. The bioleaching mechanism of Cu from printed circuit boards by *A. ferrooxidans* is speculated to be similar to that of metal sulfides [97] in terms of involving indirect leaching mediated by the biogenic sulfuric acid. The role of the microorganisms in this process is to oxidize elemental sulfur (S°) to sulfuric acid (H_2SO_4) as shown in Eq. (1). S° is not typically found in discarded PCB and added externally to the leaching medium. Ferrous iron (Fe^{2+}) is also added externally to the leaching medium, and plays the role of an electron donor. It is subsequently oxidized to ferric iron (Fe^{3+}) by bacteria (Eq. (2)).

$$S^0 + 1.5O_2 + H_2O \rightarrow 2H^+ + SO_4^{2-}\ (Microbial) \quad (1)$$

$$4Fe^{2+} + O_2 + 2H^+ \rightarrow 4Fe^{3+} + 2OH^-\ (Microbial) \quad (2)$$

In the bioleaching reaction, Fe^{3+} plays the role of an oxidizing agent for enhancing the leaching reaction as shown in Eq. (3). Biogenic ferric iron and sulfuric acid mobilizes copper from the waste material as shown in Eqs. (3) and (4), respectively. This translates into a combined acidolysis – redoxolysis bioleaching mechanism for metal dissolution from waste material.

$$Cu^0 + 2Fe^{3+} \rightarrow Cu^{2+} + 2Fe^{2+}(Chemical) \quad (3)$$

$$Cu^0 + H_2SO_4 + 0.5O_2 \rightarrow Cu^{2+} + SO_4^{2-} + H_2O\ (Chemical) \quad (4)$$

The bioleaching rate primarily depends on the initial pH, initial ferrous iron (Fe^{2+}) concentration and oxidation rate of ferrous (Fe^{2+}) to ferric ions (Fe^{3+}) [98]. Biogenic Fe^{3+} concentration is directly correlated with leaching rate (mg metal leached as a function of time) and the total extraction efficiency [28]. On the other hand, a involvement of a contact mechanism is discussed as well. *A. ferrooxidans* cells do not attach randomly to the solid surface (though chemotaxis may be involved in the preferential attachment of bacteria [46]). The interaction between *A. ferrooxidans* cells and crushed PCB particles was suspected favorable only if the van der Waals attractive force is greater than the electrostatic repulsive force, which would occur at high ionic strength of the solution only [99]. Indeed, Silva et al. [99] showed less copper being mobilized (25%) when contact was avoided. For this, ground PCB sample (particle size 500–1000 μm) were placed inside a semi-permeable membrane. Ultimately, the results for bacterial adhesion tests were found consistent with Derjaguin–Landau–Verwey–Overbeek theory [99].

Cyanide is the general term for chemicals which contain a cyano-group with the chemical formula CN^-. Trace amounts of thiocyanide (SCN) is found in human saliva, urine, and gastric juices [100]. Cyanide is suspected to be produced by bacteria under certain conditions to

create a competitive advantage for the producer microorganism and as bio-control mechanism suppressing diseases on plant roots [101,102]. Biogenic cyanide is a secondary metabolite formed by oxidative decarboxylation of glycine, as shown below in Eq. (5):

$$NH_2CH_2COOH \rightarrow HCN + CO_2 + 2H_2 \quad (5)$$

Methionine enhances the cyanide production yield but will not substitute glycine [103]. Cyanide is formed during the early stationary phase. Induction of *hcn* genes which are involved in cyanide production is initiated under conditions in which oxygen is limited, however some species can produce reasonable amounts of cyanide under normal conditions as well [104]. Biogenic cyanide production depends on several parameters, i.e., glycine concentration, initial pH, temperature, solid to liquid ratio, oxygen concentration [80,82]. Though glycine is essential for biogenic cyanide production, high concentrations have been reported to be inhibitory for growth [103,105]. Likewise, cyanide remains dissolved in solution at high pH, and gold cyanidation is reported to be most efficient in the range of 10.5–11. For some microorganisms, such high pH however, has been reported inhibitory for cell growth [106].

Microbe-REE-interactions are not yet fully understood, and bioleaching process are discussed under the terms of acidolysis, redoxolysis and complexolysis [62]. Organic acids (e.g. malic, oxalic, and citric acid) have chelating properties with some REE, donating H^+ to form metal-ligand complexes.

3.1.5. Scale-up of bioleaching applications in WEEE processing

A list of bioleaching studies involving the autotrophs and heterotrophs for critical and valuable metal recovery from WEEE is given in Table 2. Several authors investigated leaching of metals from WEEE in scaled up setups with technology readiness levels higher than 4, which translates into tests carried out in semi-pilot level. Reported metal bioleaching efficiencies were typically between 50% and 99% and between 3–7 days at solid to liquid ratio between 1–10% (*w/v*). Several studies have demonstrated improved bioleaching efficiency in sulfur- and ferrous iron-supplemented media [70,97,107]. Bottlenecks for scaling up was certainly metal bioleaching efficacy generally decreasing with increasing pulp density. Some discarded WEEE materials had an alkaline nature, and was therefore acid-consuming [94] to allow for growth of acidophiles. The non-metallic fraction, i.e. epoxy-coated substrate, organic fraction etc., of the material may be toxic to bacteria [28,108]. Further, inhibition upon direct contact of the cells with the metal-rich waste materials is a bottleneck in scaling biotechnologies [107](Xu and Ting [198]).

Ilyas et al. [69] studied bioleaching of, Al, Cu, Ni Zn from ground printed circuit boards with a moderately thermophilic acidophilic, chemolithotrophic and a heterotrophic consortium that was isolated from a local site. 64%, 86%, 74%, and 80% of Al, Cu, Ni, and Zn, respectively, was removed after a 27-day pre-leaching period followed by 280-day bioleaching period. Follow-up bioleaching studies by Ilyas et al. [97] in a reactor setup using an adapted moderately thermophilic pure culture of chemolithotrophic *Sulfobacillus thermosulfidooxidans* reached almost complete removal of Al, Cu, Zn, and Ni, at of 10% (*w/v*) pulp density. The bioleaching medium was supplemented with 25% O_2 + 0.03% CO_2, and 2.5% (*w/v*) biogenic S°, and kept at 45 °C. An interesting finding was the faster oxidation rate of biogenic sulfur over technical sulfur, which can be attributed to the higher bioavailability and hydrophilicity of biogenic S [112].

Mäkinen et al. (2015) studied the bioleaching of discarded printed circuit board (PCB) froth, using pretreatment (separation of hydrophilic / hydrophobic fractions), pre-inoculation (to favor the dominance of sulfur-oxidizers over iron oxidizers) and ultimately CSTR operation ultimately achieving copper solubilization of 99% (maximal copper concentration of 6.8 g/L).

Chen et al. [70] investigated bioleaching of copper from ground PCB using *Acidithiobacillus ferrooxidans*.Copper recovery was relatively high, (94.8% after 28 days) however, the study indicated that the rate of copper dissolution was limited by diffusion due to secondary mineral precipitation (Jarosite, iron oxyhydroxides) covering the surface of the leaching material. The formation of such jarosite precipitates may be prevented by maintaining acidic conditions (dilute sulfuric acid addition) of the leaching medium.

Compared to bioprocessing for base metal recovery, bioleaching applications related to rare earth elements are less common. Studies demonstrated the ability of selected microorganisms in bioleaching of rare earth elements (REE) from primary and secondary raw materials. Some patents on microbial REE bioleaching have been developed as well [62]. Recent studies on REE extraction through bioleaching processes are outlined in Table 3, both for primary sources and for secondary ones including electronic waste.

Ibrahim and El-Sheikh [65] investigated bioleaching of REE, Al, U, and Zn, from uraniferous gibbsite ore by Acidithiobacillus ferrooxidans in column setup, achieving 67.6% efficiency for REE (30 cycles, 0.5% elemental sulfur addition). Muravyov et al. [72] investigated REE bioleaching from metal-bearing coal ash–slag in airlift percolators, leaching Sc, Y, and La at 52%, 52.6%, and 59.5%, resp. (10 days of operation at 45 °C, 10% pulp density; 10:1 ash–slag to elemental sulfur). As mentioned above, DNI Metals operated a REE bio-heap leaching project as full scale bioleaching application [18]. Apart, to the best of our knowledge, applications at a greater technology readiness level than 6, i.e. industrial scale, involving heterotrophic microorganisms have not been reported yet.

3.2. Biosorption, bioelectrochemical and bioprecipitation processes for selective metal recovery from leachates

In the last decades, significant research efforts in the field of environmental technology were focused on the removal of toxic metals from contaminated ground waters, polluted soils and (industrial and domestic) wastewaters. These strategies include bio-based technologies, e.g. biosorption and bioprecipitation. Recently, this research focus is shifting partly towards the recovery of valuable metals from (industrial) wastewaters and polluted soils, leachates and solid wastes, including WEEE. This paradigm shift can be ascribed to the fact that bio-based technologies are considered a cost-effective option to concentrate elements from diluted wastewaters and leachates as part of these recovery strategies, and recovery of valuable elements may help to reduce the cost of waste treatment. Bio-based strategies can not only work with those dilute waste streams, but also may require a low energy input. Therefore, bioelectrochemical systems, bioprecipitation and biosorption techniques are now also being integrated into novel hydro-, bio- and hybrid-metallurgical systems.

3.2.1. Biosorption

3.2.1.1. The biosorption process. Biosorption is a physico-chemical and metabolically-independent process that includes absorption, adsorption, ion exchange, micro precipitation, surface complexation mechanisms on material of biologic origin (Volesky, 2003; [115]). Consequently, a number of materials have been studied in the frame of biosorption: living or inactive biomass of bacterial [116]; Vargas et al., 2004), fungal (Wang, and Chen [201]; Sağ [211]; Niu, and Volesky, 2007) or plant origin (Romera et al. [193]; Mack et al. [194]; Vilar et al. [195]); agricultural residues, waste crustacean biomass, etc. (Volesky [212]; Wang and Chen [199]). New members of biosorbent family are waste materials or biomass by-products from large scale fermentation processes (Kapoor and Viraraghaven [200]; Wang and Chen, 2006). Fig. 3 displays a schematic classification of the biosorption mechanisms [117].

The efficiency of metal biosorption is affected by the structure of the biosorbent, especially cell surface and the cell wall (Volesky [212]; Wang and Chen, [199]; 186,118]). Several functional groups are involved, such as carboxyl, imidazole, sulfhydryl, amino, phosphate,

Table 2
Recent studies on biotechnological strategies for valuable metal recovery from WEEE, operational conditions and metal yields.

Microorganisms	Operational parameters			Bioleaching mechanism	Leached metals % (mg/g PCB)	References
	T(°C)	pH	Pulp density (S:L[a])			
Technology readiness level < 4 (batch reactors)						
Autotrophic bioleaching						
Sulfobacillus thermosulfidooxidans, acidophilic isolate	45 °C	2.0	10:1000	Acidolysis (H_2SO_4), Redoxolysis (Fe^{3+})	Cu 89% (76 mg/g), Ni 81% (16.2 mg/g), Zn 83% (66.4 mg/g)	[69]
Acidithiobacillus sp., Gallionella sp., Leptospirillum sp.	30 °C	1.5 - 2.5	20:1000	Redoxolysis (Fe^{3+})	Cu 95% (219 mg/g)	[98]
At. ferrooxidans, At. thiooxidans	28 °C	1.5 - 3.5	30:1000	*Acidolysis (H_2SO_4), redoxolysis (Fe^{3+})*	Cu (94%), Ni (89%), Zn (90%)	[107]
Acidophilic consortium (genera Acidithiobacillus and Gallionella)	30 °C	2.0	12:1000	Redoxolysis (Fe^{3+})	Cu 97% (626 mg/g), Al 88% (34 mg/g), Zn 92% (28 mg/g)	[28]
At. ferrooxidans, Leptospirillum ferrooxidans, At. thiooxidans	25 °C	1.7	10:1000	Acidolysis (H_2SO_4), redoxolysis (Fe^{3+})	Cu 95% (106 mg/g)	[61]
Acidithiobacillus thiooxidans	30 °C	0.5	10:1000	Acidolysis (H_2SO_4)	Cu 98% (132 mg/g)	[67]
At. caldus, Le. ferriphilum, Sulfobacillus benefaciens, Ferroplasma acidiphilum	37 °C	1.7	10:1000	Acidolysis (H_2SO_4), redoxolysis (Fe^{3+})	Cu 99% (29 mg/g)	[109]
Heterotrophic leaching						
Aspergillus niger, Penicillium simplicissimum	30 °C	3.5	10:1000	Acidolysis (organic acids)	Cu 65% (52 mg/g), Al 95% (225 mg/g), Ni 95% (14 mg/g), Zn 95% (25 mg/g)	[94]
Chromobacterium violaceum, Pseudomonas fluorescens, Pseudomonas plecoglossicida	30 °C	7.2 - 9.2	various	Complexolysis (CN^-)	Au 69% (not specified)	[51]
Chromobacterium violaceum (metabolically engineered)	30 °C	Neutral	5:1000	Complexolysis (CN^-)	*Au 31% (0.04 mg/g)*	[110]
Ps. chlororaphis	25 °C	7.0	19:1000	Complexolysis (CN^-)	Au (8%), Ag (12%), Cu (52%)	[52]
At. ferrooxidans, At. thiooxidans, Thiobacillus denitrificans, Thiobacillus thioparus, Bacillus subtilis, Bacillus cereus	22 - 37 °C	5.0 - 7.0	10:1000	Acidolysis (H_2SO_4), redoxolysis (Fe^{3+}), complexolysis (surfactants)	Cu 53% (22 mg/g), Ni 48.5% (6.4 mg/g), Zn 48% (6 mg/g)	[111]
Ps. putida (two-step)	30 °C	8.0 - 9.2	10:1000	Complexolysis (CN^-)	Cu 98% (164 mg/g), Au 44% (0.1 mg/g)	[71]
Technology readiness level > 4 (column and tank reactors)						
Autotrophic bioleaching						
Sb. thermosulfidooxidans, Thermoplasma acidophilum	45 °C, 1.5-2.7, n/a (10 kg)			Acidolysis (H_2SO_4), redoxolysis (Fe^{3+})	Cu 86% (76 mg/g), Zn 80% (71 mg/g), Ni 74% (15 mg/g), Al 64% (6.5 mg/g)	[97]
Sb. thermosulfidooxidans	45 °C, 2.0, 1.5-3.5%			Acidolysis (H_2SO_4), redoxolysis (Fe^{3+})	Cu 95% (105 mg/g), Al 91% (19 mg/g), Zn 96% (18 mg/g), Ni 94% (18 mg/g)	[112]
At. ferrooxidans	30 °C, 2.0, 1.5%			Acidolysis (H_2SO_4), redoxolysis (Fe^{3+})	Cu 97% (247 mg/g), Zn 84% (52 mg/g), Al 75% (47 mg/g)	[113]
At. ferrooxidans, At. thiooxidans	28 °C, 1.1-1.6, 1%			Acidolysis (H_2SO_4), redoxolysis (Fe^{3+})	Cu 99% (151 mg/g)	[114]
At. ferrooxidans	30 °C, 2.0, 1%			Acidolysis (H_2SO_4), redoxolysis (Fe^{3+})	Cu 95% (203 mg/g)	[70]

[a] S/L: Solid to liquid ratio.

Table 3
Recent studies on biotechnological strategies for REE extraction from solid matrices.

Microorganism(s)	Matrix	Operational parameters			Bioleaching mechanism	Leached metals (%)	References
		T(°C)	pH	S/L[a] (%, *w/v*)			
Technology readiness level < 4 (batch reactors)							
Autotrophic bioleaching							
Acidithiobacillus ferrooxidans, Acidithiobacillus thiooxidans, Leptospirillum ferrooxidans	CRT fluorescent powder	30 °C	n/a	10%	Acidolysis (H_2SO_4), redoxolysis ($Fe3^+$)	Y 70%	[68]
Heterotrophic leaching							
Aspergillus ficuum	Monazite mineral	30 °C	3	0.6%	Acidolysis (organic acids), Complexolysis	Total REEs 75.4%	[88]
Pseudomonas aeruginosa	Monazite mineral	35 °C	6	0.6%	Acidolysis (organic acids)	Total REEs 63.5%	[88]
Penicillium tricolor	red mud	30 °C	2-2.5	2%	Acidolysis (organic acids)	La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Y, Sc 36 – 78%	[73]
Acetobacter aceti	Monazite mineral	30 °C	3-5	16.6%	Acidolysis (organic acids), Complexolysis	Ce 0.13%, La 0.11%	[89]
Aspergillus ficuum	thorium-uranium concentrate	25 °C	3	0.75 %	Acidolysis (organic acids)	La 20%, Ce 33%, Y 2.5%	[64]
Aspergillus niger, Aspergillus terreus, Paecilomyces	Monazite mineral	25–28 °C	2-2.8	1%	Acidolysis (organic acids)	Ce, La, Nd, Pr 3-5%	[63]
Gluconobacter oxydans	fluorescent lamp phosphor powder	30 °C	n/a	1.5%	Acidolysis (organic acids)	Total REEs (Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Ho, Dy, Er, Tm, Yb, Lu, Th) 2%	[74]
Technology readiness level > 4 (column and tank reactors)							
Autotrophic bioleaching							
Acidithiobacillus ferrooxidans	Gibbsite mineral	25 °C	2	1kg/500mL	Acidolysis (H_2SO_4)	REEs 67.6%	[65]
Acidophilic chemolithotrophs	Coal ashes	45 °C	0.9- 2	10%	Acidolysis (H_2SO_4)	Sc 52%, Y 52.6%, La 59.5%	[72]

[a] S/L: Solid to liquid ratio.

sulfate, thioether, phenol, carbonyl, amide and hydroxyl moieties are involved in the sorption processes (Wang and Chen [199]; Volesky, 2007). It has been proven that a higher sorption capacity can be achieved by functionalizing non-living biomaterials with chelating agents, or by other chemical (e.g., alteration of functional groups or use of crosslinking agents) or physical (e.g., heat treatment, ultrasonic treatment) modification of the materials (Fig. 3). For instance, Ru loading (from 86 to 145 mg/g dry cells) and selectivity over Ni and Zn were improved by the acid pretreatment of *Rhodopseudomonas palustris* cells, whereas the Ru loading capacity of the biomass was increased by 6.9-fold using chitosan and surface modification with Polyethyleimine (PEI) [119,120]. Recently, some novel chemically modified biosorbents have been developed that show excellent selectivity to recover valuable metals from industrial wastewaters and leachates of industrial residues, generating pure metal solutions (e.g., Roosen and Binnemans [202]; Roosen et al., 2016). However, functionalization / pretreatment often increase the cost and environmental impact of the technology considerably [81,115].

Further, the efficiency of biosorption is related to the surface area of the biomass [118], with cellular microorganisms exhibiting the highest biosorption potential. Efficacy of the (de)sorption processes is affected by (1) the form in which metals are present, i.e. their speciation [73], and (2) operational parameters (e.g., temperature, biosorbent dosage and pH) and (3) the type of sorbent used [118,121,122].

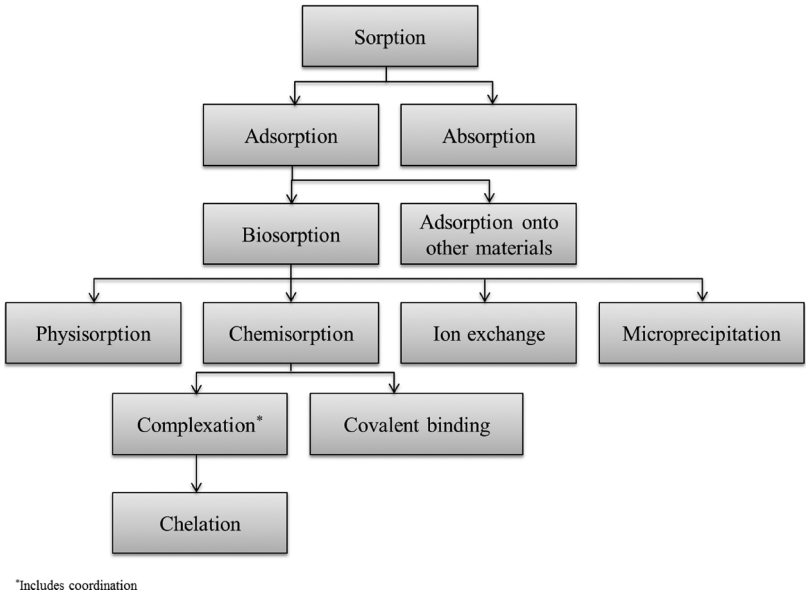

Fig. 3. Schematic explanation of metal biosorption mechanisms (Redrawn after [117]).

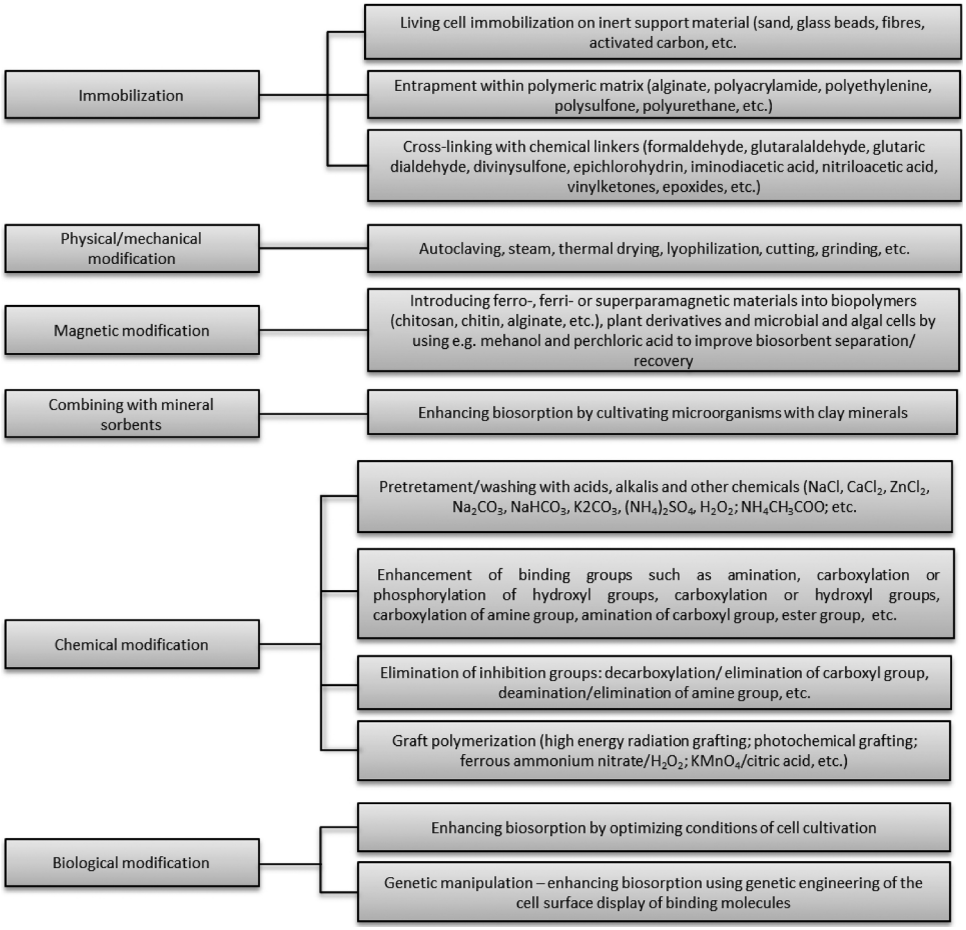

Fig. 4. Biosorbent pre-treatment and modification methods to improve the uptake capacity (redrawn after [115]).

To summarize, biosorption has the following advantages: (1) the elements can be absorbed selectively even at very low concentrations, (2) the process is energy efficient, (3) the process conditions are mild in terms of e.g. pH and temperature, (4) the elements can be harvested easily, and (5) some biosorbents can be regenerated if needed (Kücüker et al. [186]). Despite the research efforts since 1990's, there has been limited exploitation of biosorption in an industrial context [81] in general, and of biosorption-based recovery of metals from WEEE leachate solutions in particular. The lack of commercial success is attributed by Fomina and Gadd [115] still poor understanding of the mechanisms, kinetics and thermodynamics of the processes, as well as the wide range of competing technologies that are available. In addition, further drawbacks for biosorption particular relating to WEEE are (1) lack of data allowing to assess their selectivity for complex, highly acidic / multi metal rich waste streams different from the original tested solutions, (2) that chemical modifications of the materials are often needed to increase their selectivity, and (3) the need to initially dissolve metal ions from a complex solid matrix (Fig. 4).

3.2.1.2. Recovery of metals by biosorption. Table 4 gives a list of studies on biosorption of REEs and PGMs (Wang and Chen [199]; [74]; Kücüker et al. [186]). These studies mainly focused on the application of biosorption processes to recover metals from WEEE leachates. Studies on recovery from wastewaters generated by WEEE industries are still scarce. For instance, Bhat et al. [117] have proposed an integrated model for the recovery of Au and Ag from WEEE using a combination of hydrometallurgical and biometallurgical processes. They concluded that *Eicchornia* root biomass and waste tea powder were efficient biosorbents for recovery of leached silver-cyanide from electronic scrap, and the concentrated silver-cyanide recovered in the biosorption process could further be used as an input material for electroplating industry. Côrtes et al. [33] studied biosorption of gold alone and in combination with precipitation from discarded computer microprocessor (DCM) thiourea leachates using chitin as a biosorbent. Ultimately, about 80% of the gold were recovered at 20 g/L of chitin within 4 h.

Kücüker et al. [80] studied Nd removal from neodymium magnet leachates in batch and continuous sorption systems by using dried green microalgae (*Chlorella vulgaris*). The maximum Nd uptake (q = 157.21 mg/g sorbent) was determined at pH 5 with a biosorbent dosage of 500 mg/L and an initial neodymium concentration in the mixed leachate solution of 250 mg/L at 35 °C. Though *Chlorella vulgaris* was considered a potent biosorbent, substantial pH adaptation was needed prior to biosorption, which may be a bottleneck. Due to the above mentioned bottlenecks, most biosorption processes are still at the laboratory scale. Still, biosorption may find applications in the recovery of valuable metals. Furthermore, the use of hybrid technologies for selective metal recovery and the potential of the technology to directly produce valuable products, including e.g. micronutrient-enriched feed supplements and fertilizers, from waste streams should also be explored [115,121,122].

Most of the previous studies on metal biosorption focused on biosorption for pollutant removal and, therefore, recovery of loaded metals from the waste streams was not even considered. In particular selective desorption from the biosorbent may proof a challenge [81,115], in particular since most previous studies focused on metal sorption in lab-scale using synthetic solutions.

3.2.2. Bioprecipitation and bioelectrochemical systems (BES)

3.2.2.1. Bioprecipitation and bioreduction. Bioprecipitation processes to

Table 4
Recovery of critical metals from aqueous sources through biosorption.

Cation	Biosorbent	Biosorption capacity (mg/g)	References
Ag^{+}	*Chemically modified chitosan resin*	413.62	Donia et al. [184]
	Bacillus cereus	91.75	Li et al. [14]
	Saccharomyces cerevisiae	135.91	Chen et al. [46]
	Klebsiella sp.3S1	141.1	Muñoz et al. [18]
	Magnetospirillum gryphiswaldense	13.5	Wang et al. [123]
Au^{3+}	*Fucus vesiculosus*	68.94	Mata et al. (2009)
	Rice husk carbon	1496.90	Chand et al. (2009)
	Chemically modified chitosan	669.66	Donia et al. [184]
	Crosslinked chitosan resin	70.34	Fujiwara et al. (2007)
	Silk and chitosan	0.20	Chen et al. (2010)
	Thiourea modified alginate	1668.25	Gao et al. [52]
Pd^{2+}	*Racomitrium lanuginosum*	37.2	Sari and Mustafa et al. [185]
	Bayberry tannin	33.4	Ma et al. [124]
Y^{3+}	NaOH modified *Pleurotus ostreatus*	45.45	Hussein et al. [28]
La^{3+}	Fish scales	250.00	Das and Varshini [176]
	Pleurotus ostreatus basidiocarps	54.54	Hussien [214]
	Chlamydomonas reinhardtii	142.86	Birungi and Chirwa [25]
	Sargassum sp.	91.68	Oliveira and Garcia [125]
	Chlorella vulgaris	74.60	Birungi and Chirwa [25]
Ce^{3+}	Grapefruit peel	159.30	Torab et al. (2015)
	Prawn carapace	1000.00	Varshini and Das [176]
	Fish scales	200.00	Varshini and Das [176]
	Corn style	250.00	Varshini and Das [176]
	Platanus orientalis	32.05	Sert et al. [126]
Pr^{3+}	*Green seaweed (Ulva lactuca)*	69.75	Vijayaraghavan [215]
	Free Turbinaria conoides (brown seaweed)	146.4	Vijayaraghavan and Jegan [177]
	Polysulfone immobilized Turbinaria conoides	119.5	Vijayaraghavan and Jegan [177]
	Crab shell	66.60	Varshini and Das (2015b)
	Sargassum sp.	98.63	Oliveria et al. (2011)
Nd^{3+}	*Chlorella vulgaris*	157.21	Kücüker et al. [186]
	Physcomitrella patens	106.73	Heilmann et al. [126]
	Calothrix brevissima	69.23	Heilmann et al. [126]
	Tetraselmis chuii	51.92	Heilmann et al. [126]
	Sargassum sp.	100.96	Oliveira and Garcia [125]
Sm^{3+}	Activated biochars from cactus fibres (pH = 3.0)	90.00	Hadjittoti et al. (2016)
	Activated biochars from cactus fibres (pH = 6.5)	350.00	Hadjittoti et al. (2016)
	Sargassum sp.	105.25	Oliveria et al. (2011)
Eu^{3+}	*Activated carbon*	86.00	Anagnostopoulos and Asaymeopoulos (2013)
	Modified cactus fibres (MnO2-coated))	0.46	Prodromou and Pashalidis [178]
	Sargassum polycystum Ca-loaded biomass	62.30	Diniz and Volesky [79]
	Sargassum polycystum Ca-loaded biomass	62.30	Oliveira and Garcia [125]
Yb	*Sargassum polycystum* Ca-loaded biomass	48.45	Diniz and Volesky [79]

remove metals from (industrial) waste streams typically have focused on the use of sulphate reducing bacteria, forming insoluble metal sulphidic precipitates, as illustrated in the review published by Hussain et al. [89]. Sulphate reduction processes have mainly been used in mining and metal recycling industries. Furthermore, metabolizing bacteria can release other compounds which can be used in precipitation processes (e.g., phosphates) and/or induce changes of the aqueous environment, such as pH changes, which may promote precipitation [121]. Janyasuthiwong et al. [26] successfully applied sulphate reducing bacteria to precipitate Cu from PCB leachates (1.0 M HNO_3) in a continuous system, yielding more than 90% Cu recovery.

Furthermore, direct bioreduction of precious metals to metallic nanoparticles also has received attention in recent years. Production of nanomaterials from waste materials using microorganisms is particularly promising because (1) the produced nanomaterials can be directly valorized in catalytic/industrial processes, (2) the microorganisms are considered as inexpensive catalysts and synthesis is conducted at ambient temperature and pressure. Several researchers have focused on studying Pd bioreduction in the form of biomass-associated nanopalladium (Pd(0) or bio-Pd) [127]. The microbial activity seems particularly useful for in situ generation of reducing agents, such as hydrogen, for Pd(II) reduction as well as for the subsequent catalytic reactions [15,110,128,129]. For instance, microbial reduction of Pd(II) to bio-Pd was used to recover the element from industrial automotive catalyst, and from the leachates of catalytic transformation of chemicals and pollutants applications [86]. A three-stage biobased process using native and palladized D. desulfuricans was also already developed for selective sequential recovery of Au and PGMs from e-waste leachates [21].

Formation of other biogenic metallic (nano)particles was also previously investigated and shown to have potential to generate products from waste streams which can be directly valorized. For example, Deplanche et al. [37] focused on biorecovery of gold from jewellery wastes by *Escherichia coli* and biomanufacture of catalytically active Au-nanomaterials for the oxidation of glycerol. Work of [22,23] exemplified the fabrication of novel gold nanostructures and stable bio-Au nanocomposites with excellent optical properties by combining microorganisms and a surfactant. De Gusseme et al. [179,180] illustrated the potential use of biogenic Ce and Ag(0) particles for virus disinfection, whereas [170,171] have prepared membranes containing biogenic Ag (0) precipitates having antifouling properties. The bio-Ag-0/PES composite membranes, even with the lowest content of biogenic silver (140 mg bio-Ag(0) m^2), not only exhibited excellent anti-bacterial activity, but also prevented bacterial attachment to the membrane surface and decreased the biofilm formation. De Corte et al. [93] have developed biosupported bimetallic Pd-Au nanocatalysts which can be used for dechlorination of environmental contaminants. Although all of this work is very promising, many of these studies still used synthetic media to prepare the biogenic particles and illustrate their potential for removal and recovery of valuable metals from waste. Therefore, there is

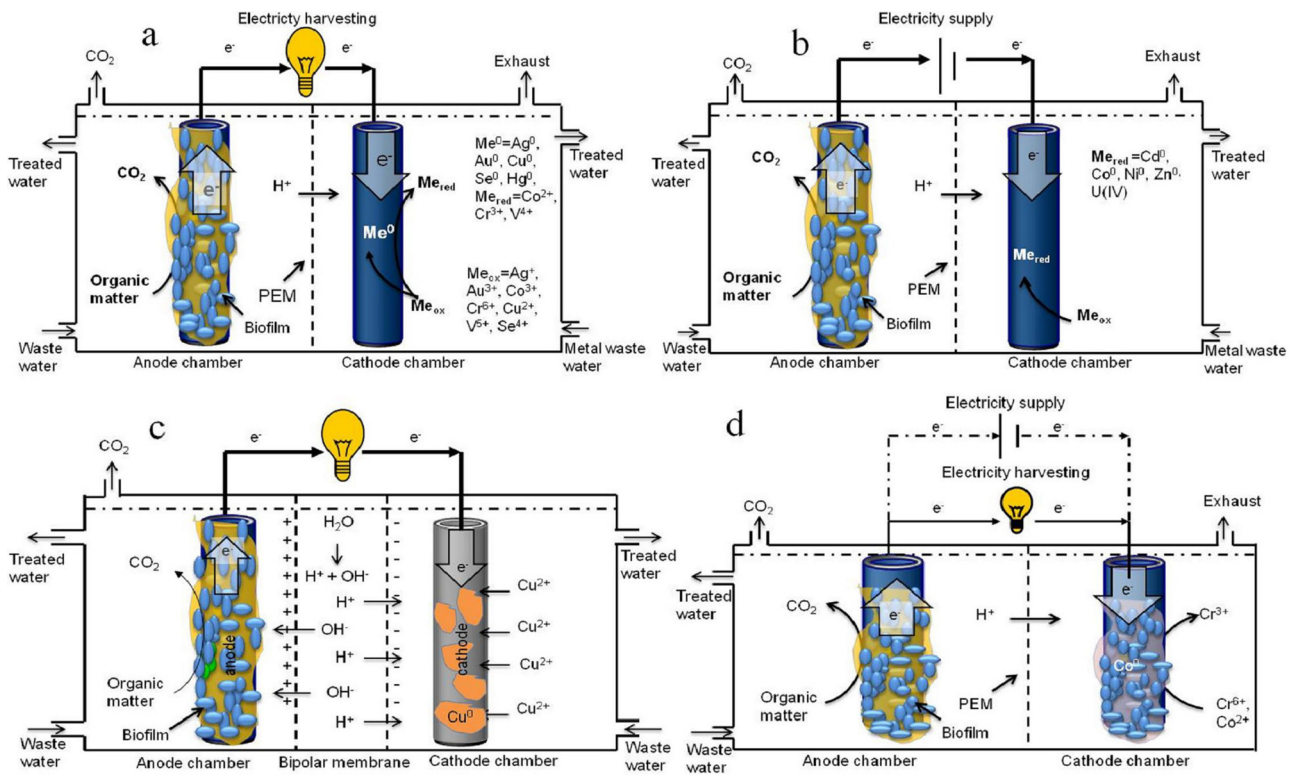

Fig. 5. Removal and recovery of heavy metals in (a) microbial fuel cells, (b) microbial electrolysis cells, (c) microbial fuel cell with bipolar membrane (modified and redrawn after ter Heijne et al. [182] and (d) microbial fuel cells and microbial electrochemical cells with biocathodes.

currently a clear need for setting up experiments focused on the generation of biogenic particles from real waste streams, including WEEE, and upscaling of the technologies to pilot-scale.

3.2.2.2. Bioelectrochemical systems (BES). Bioelectrochemical systems (BES) link electrochemical systems with the microbial metabolism. Microorganisms can respire on electrodes, harvesting electrons from waste streams containing organic matter. This can be used to generate electricity (in microbial fuel cells - MFCs), treat contaminants and/or produce chemical products, such as methane, ethanol, or hydrogen peroxide (in microbial electrolysis cells - MECs) (Fig. 5). However, the electrons removed may also be used to recover metals from solution at the cathode. Metals can be reduced at the cathode with a net positive cell potential and power generation if the potential generated at the bioanode is lower than the redox potential of the half-cell reaction at the cathode, which is often the case. In other cases, supply of voltage is needed. Recovery of several base metals (e.g., Cu, Ni, Cd, Zn) as well as of a few precious and scarce metal(loid)s (e.g., Ag, Au, Co, and Se) have recently been demonstrated in BESs (e.g., [78,130,59]). Most researchers worked with synthetic solutions, delivering a proof-of-principle. However, Peiravi et al. [54] and Pozo et al. [131] recently designed bioelectrochemical systems for treatment of real mine drainage. Pozo et al. [131] produced a solid metal sludge which was twice less voluminous and 9 times more readily settleable than metal-sludge precipitated using NaOH. Concomitant precipitation of rare earth elements, among other high-value metals, occurred, which could be used to offset the treatment costs. Additional studies are still needed to investigate the possibility to recover metals from other real waste streams and leachates, e.g. those generated by WEEE processing, using BES and to scale up the technology. Furthermore, the potential application of the BES platform for recovery of a range of other precious and technology-critical metals, such as metals occurring in WEEE, can still be further explored.

4. Perspectives and future developments

Global waste electrical and electronic equipment (WEEE) generation will increase in the next years, particularly in the developing countries. WEEE is an important secondary source of critical raw materials. These metals play a central role particularly in the transition to a green society. Their secure supply is under risk, therefore alternative supply sources, for instance metal-rich post-consumer waste materials, are of importance. However, the potential economic benefits from critical material recovery should not be perceived as the sole driver to develop these technologies. Improper disposal / handling of WEEE and many other toxic metal-bearing secondary sources, is a risk for the environment and public health. Admittedly, it is challenging to assign an economic value to a risk that has been mitigated. Environmental sustainability accounting tools, e.g. Life Cycle Assessment (LCA) and Life Cycle Costing (LCC) might prove useful to map the environmental hotspots and to communicate this to the public and the decision makers. Biotechnology is highlighted to play a significant role in the treatment and resource recovery from metal-containing waste materials [13,20,43]. Bioprocessing of waste for metal recovery attracts interest to meet two objectives: resource recovery and pollution mitigation. Biotechnology will provide a number of technological innovations supplementing conventional technologies in recovery and re-use of critical metals from secondary sources in the transition to a sustainable management of WEEE.

Biotechnologies have a historical niche area in processing of low-grade ores. WEEE is distinct compared to primary ores in terms of its chemical composition, abundance of the metals and their complexity. WEEE typically include high concentrations of conventional metals and a lower concentration of critical metals in various mixtures. Current WEEE recycling practices are inadequate to target the critical metals, which are typically found in low concentrations [22]. It is important to note that conventional (established) bioleaching operations make use of autotrophic microorganisms that can conserve energy from solubilizing

(oxidizing) sulfidic ores. WEEE, on the other hand, include metals in their native metallic form. Thus, it is required to supplement the microbes with additional energy source. This specific challenge requires novel strategies for critical metal recovery from WEEE.

There is need to perform more fundamental research on WEEE bioprocessing as some of the main leaching mechanisms are not fully understood. As for metals present in other forms (carbonates, oxides or silicates), the principles / experiences using autotrophic bioleaching cannot simply be transferred to WEEE bioleaching due to the fundamentally different underlying chemistry (i.e. WEEE not containing metals in form of sulfides). Further investigations are required in order to advance further into full-scale applications, including optimization of the operational conditions and assessment of environmental impacts is needed. In addition, including scale-up studies with techno-economic assessment and environmental sustainability analysis considerations are important considerations in biotechnological strategies for metal recovery.

Acknowledgements

The authors would like to acknowledge networking support by the COST Action ES1407 funded by the European Commission (EC). Ata Akcil acknowledges the Scientific and Technological Research Council of Turkey for the INTENC Project (113Y011) during 2014-2018.

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