Integration of Bioleaching and Biorefinery Technologies for the Recovery of Base and Critical Elements from Electronic Waste

Alexandre H. Kompalitch and Eric D. van Hullebusch

Abstract

Several profound societal changes such as the shift towards renewable energies have created an ever-increasing demand for base and critical metals. Electronic wastes constitute a significant secondary source of such elements and a potential environmental hazard if disposed of improperly. In contrast to traditional methods of recycling e-waste, biohydrometallurgy is an environmentally friendly, low-cost, and energy-efficient alternative. Although processes developed in laboratories display promising yields, it is still premature to implement these biotechnological strategies on a larger scale as the bioleaching and biorefinery mechanisms are still poorly understood. Moreover, very few studies focus on fully biological processes, and most opt for more efficient hybrid approaches. Thus, this book chapter compiles the optimal parameters reported in recent studies, from waste pre-treatment to metal biorecovery, along with insights to complete and close the biohydrometallurgical recycling loop.

Keywords

Biohydrometallurgy · Bioleaching · Biorecovery · E-waste · Pregnant leaching solution · Process optimization · Critical metals

1 Introduction

The advent of advanced technologies, increasing urbanization, and global economic development are the main causes for the exponential production and consumption of electrical and electronic equipment (EEE). It ranges from small day-to-day products, such as monitors, personal computers, cell phones, microwaves, and lamps, to large household products, namely refrigerators, electric stoves, washing machines, and air conditioners. Programmed obsolescence and constant technological innovation, combined with increasing consumer demand for affordable equipment and high living standards, drastically reduce these products' lifespan. These phenomena lead to an unprecedented production of waste (Tansel, 2017), more commonly known as waste electrical and electronic equipment (WEEE or E-waste). E-waste management and recycling are a real challenge but also an opportunity for our modern societies. E-waste recycling represents a non-negligible source of metals. The average e-waste contains around 60% of metals and metalloids (Gopikrishnan et al., 2020), ranging from rare earth elements (REE) to platinum group metals (PGM), that are essential for the production of EEE and the development of low-carbon energies.

E-waste is the most prevalent and fastest-growing source of waste (Darby & Obara, 2005). In 2019, 53.6 Mt of e-waste was globally generated, but only 17.4% (9.3 Mt) is documented to be collected and recycled. E-waste production is estimated to reach 74.7 Mt by 2030, an increase of 39% (Forti et al., 2020). The huge amount of e-waste generated each year represents a serious threat to the environment when improperly managed. It is usually discarded in landfills or transferred to developing countries. E-waste contains toxic substances such as arsenic, lead, cadmium, mercury, and plastics that can leach into the soil, contaminate the environment and therefore the human habitat (Widmer et al., 2005). Although 78 countries have adopted legislation on e-waste treatment, most have insufficient policies or poor enforcement. In 2019, Europe was the continent with the most functional e-waste policy, achieving a collection and recycling rate of 42.5%. It was followed by Asia (11.7%), Americas (9.4%), Oceania (8.8%), and Africa (0.9%) (Forti et al., 2020).

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Fig. 1 Schematic representation of a biohydrometallurgical process cycle as conceptualized in this review
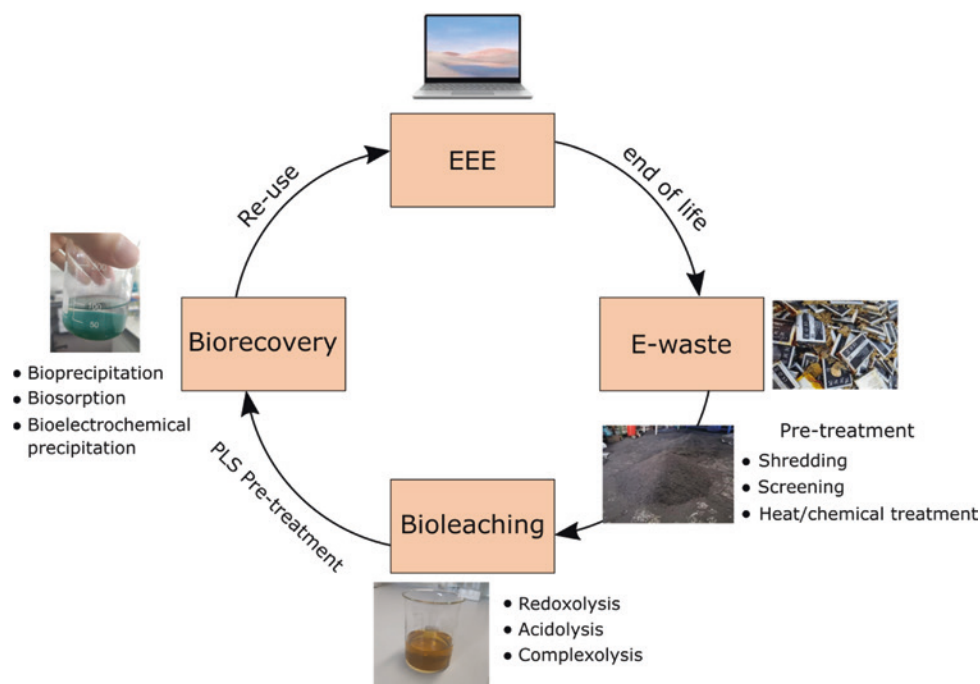
Currently, e-waste is industrially recycled through pyrometallurgical and hydrometallurgical processes. Both routes have major drawbacks. The former approach consists of smelting waste at high temperatures of up to 900 °C, is energy-intensive, lacks selectivity, and involves loss of some metals (Becci et al., 2020; Sethurajan & Gaydardzhiev, 2021). The main issues related to the hydrometallurgical route are its intensive use of toxic chemicals causing environmental problems, the high costs, and the production of hazardous substances (Mishra et al., 2021). Biohydrometallurgy uses microorganisms in an aqueous medium to recover metals from various sources (Kaksonen et al., 2018). It is considered as an environmentally friendly and sustainable alternative to established e-waste treatment and metal recovery processes. It allows for simpler, less energy-intensive, and cost-effective processes (Ilyas et al., 2014). These techniques are already being used successfully for mining primary and secondary ores (e.g. biomining, heap bioleaching) (Jerez, 2017; Johnson & Du Plessis, 2015; Thenepalli et al., 2019) and are extremely promising for e-waste recycling. Research on the bioprocessing of e-waste for the recovery of critical metals is at an early stage but is growing rapidly. It generally involves two steps in which microorganisms have central roles in the transformation processes. The first is bioleaching, which is the conversion of metals from their solid to their soluble form through the bioproduction of leaching compounds or metabolites. Biorecovery is the second step and allows the bioprecipitation of dissolved metals that could be selectively recovered in some specific cases.

Very few studies related to biohydrometallurgy fully combine bioleaching and bioprecipitation steps (e.g.

Sethurajan et al., 2017). These two processes are often coupled with hydrometallurgy, i.e. either biorecovery preceded by chemical leaching (or the use of a synthetic pregnant leaching solution) (Calvert et al., 2019; Creamer et al., 2006; Kim et al., 2018) or bioleaching followed by chemical precipitation (Ijadi Bajestani et al., 2014). Moreover, most biorecovery studies are focused on acid mine drainage and wastewater treatment, as well as primary ore biomining. Therefore, there is a clear need to integrate real pregnant bioleaching solutions (PLSs) coming from e-waste with biorecovery setups to complete the biohydrometallurgy recycling loop (Fig. 1).

This book chapter attempts to reflect on the means and critical parameters to be considered to achieve this. To this end, various up-to-date studies on the bioprocessing of e-waste and the biorecovery of base and critical metals have been reviewed. First of all, pre-treatment of the three types of e-waste that have been most frequently found in recent biometallurgical studies are documented: printed circuit boards, liquid–crystal displays, and lithium-ion batteries. Several bioleaching and biorecovery parameters are then examined to determine which are most critical for the successful integration of bioleaching for biorecovery technologies.

2 Electronic Waste Pre-treatment

Pre-treatment of e-waste is a crucial step that must be taken prior to metal recovery in order to optimize conditions for biohydrometallurgical processes. Depending on the type

of waste treated, the structure of electronic scrap can be extremely complex due to the manufacturing process and the diversity of the components used.

The pre-treatment of lithium-ion batteries, printed circuit boards, and liquid–crystal displays are presented, as these are the WEEEs that are the most commonly treated in recent bioleaching and bioprecipitation studies.

2.1 Printed Circuit Boards

Printed circuit boards (PCBs) are the basic building block of all electrical and electronic equipment since they support and connect all other components. PCBs represent 4–7 wt% of the total EEE generated and 2–3% of the global amount of E-waste generated (Dalrymple et al., 2007; Zhou & Qiu, 2010).

PCBs' most common base material is FR-4, which is a non-conductive composite material made of epoxy resin reinforced with woven fibreglass cloth. Because of its conductive properties, copper foil laminate is either printed or engraved on and inside the non-conductive substrate. Electric components such as diodes, capacitors, and transistors are then mounted and soldered on the substrate.

Generally, PCBs can contain up to 40% metals, 30% plastics, and 30% glass and ceramics (Kang et al., 2021; Ribeiro et al., 2019). The glass substrate is mainly composed of SiO_2, CaO, Al_2O_3, MgO, and BeO (Ghosh et al., 2015). Copper constitutes about 10–20% of PCBs as it is the main conductive material, but other metals are like Au, Ag, Pd, Pb, Sn, Sb, Ni, Al, Be, and Cd are also found. It has been reported that in PCB, Au and Ag concentrations can be as high as 700 g/t and 800 g/t, respectively (Kasper & Veit, 2018; Ribeiro et al., 2019). The concentration of Cu in waste PCBs is 13–26 times higher than in primary ores and that of Au is up to 50 times higher (Akcil et al., 2015; Cui & Zhang, 2008; Zhang & Forssberg, 1998). PCBs are commonly treated with brominated flame retardants (BFRs) such as polybrominated diphenyl ethers (PBDEs), tetrabromobisphenol A (TBBP-A), and hexabromocyclododecane (HBCD) (Alaee & Wenning, 2002). When incinerated, these compounds can react with the oxygen present in the atmosphere to form dioxins and furans (e.g. polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs), polybromochlorodibenzo-p-dioxins and dibenzofurans (PXDD/Fs)) which are highly toxic for the human body (Wong et al., 2007).

Dismantling is the first step in waste PCB pre-treatment. Electrical components are dismounted and treated separately. This process can be done manually or automatically by applying pressure and destroying the solders, composed of a 63–37% Sn–Pb alloy. Although melting the solders has also been reported as an efficient way of dismounting ECs

(Yokoyama & Iji, 1997), it can also produce enough heat to release dioxins. The laminated structure of PCB waste requires a reduction in particle size to increase the contact surface between the metals and the leachate. Industries employ several hammer crushers, rotary crushers, and disc crushers, but shredding and cutting remain the most effective methods (Ghosh et al., 2015).

2.2 Lithium-Ion Batteries

Lithium-ion batteries (LIBs) are nowadays the most common energy storage device because they are manufactured in products such as smartphones and laptops that have become part of our everyday life. They also play a decisive role in the energy transition as they are a central element in the construction of electric vehicles, due to their lightweight, high-energy capacity per unit of weight, and relatively long lifespan of up to 5 years (Mylarappa et al., 2017). Hence, spent LIBs potentially represent a very large waste stream. More than one million electric vehicles were sold globally in 2017, which could represent ultimately almost 250,000 tonnes of battery waste (Lie et al., 2020; Sethurajan & Gaydardzhiev, 2021).

Spent LIBs are a significant source of critical metals. LIBs function by exchanging lithium ions between a cathode and an anode via an electrolyte to generate electricity. Generally, the anode is composed of graphite, a polyvinylidene fluoride (PVDF) binder, and the cathode of lithium cobalt dioxide ($LiCoO_2$). Other lithium transition metal oxides such as LiV_2O_3, $LiMn_2O_4$, $LiNiO_2$, $LiFePO_4$, and $Li[Ni_xMn_yCo_z]O_2$ are less popular than $LiCoO_2$ because of their lower energy density and lifespan. Each anode and cathode are surrounded by current collectors, made of aluminium and copper foil, and the surrounding casing is made of iron.

The first step in the pre-treatment of spent LIBs is discharging. This step is essential because dismantling can cause short circuits or spontaneous combustion (Zheng et al., 2018). This is usually done by shorting the battery under a rolling press and then immersing it in a salt solution. One of the issues encountered in this process is the leakage of some of the internal components of the battery into the solution. Salts such as NaCl and KCl contain chloride ions that can corrode the iron casing. It has been reported that the use of $FeSO_4$ can provide a good compromise between discharge performance and leakage limitation (Kim et al., 2021; Xiao et al., 2020). The next step is to separate the electrodes from the other parts of the battery. Different methods are used such as heat treatment, organic solvent, and alkaline dissolution, but mechanical methods such as sieving and wet or dry crushing seem to prevail for bioleaching studies, as particle size reduction is a

crucial parameter affecting leaching efficiency. Studies generally aim to achieve a fine particle size of around 75 μm (Heydarian et al., 2018; Horeh et al., 2016), but coarser particles are also used (Biswal et al., 2018; Roy et al., 2021).

2.3 Liquid–Crystal Displays

Liquid–crystal displays (LCDs) are flat panel displays that use the optical properties of liquid crystals combined with a polarizer. LCD production has regularly exceeded 200 million units since 2010 (Zhang et al., 2015). In addition, the lifetime of an LCD panel is typically 3–5 years. End-of-life LCDs can therefore be considered as a non-negligible secondary metal resource.

LCD monitors are constituted of an LCD panel, printed circuit boards (PCBs), and a metal frame. The LCD panel features a glass substrate and a backlight unit, which can account for 15–40 wt% each (Li et al., 2009). Spent LCDs as a secondary resource are mostly interesting for the recovery of indium, a post-transition metal used in the manufacture of optoelectronic and semiconductor compounds. About 70% of the indium is found in indium tin oxide (ITO) film, an In–Sn alloy composed of indium oxide (In_2O_3) and tin oxide (SnO_2) (Zhang et al., 2015). Indium is generally reported to be present in ITO at a concentration of 102 mg/kg (Li et al., 2009).

To recover indium from LCDs, the ITO film contained in the glass panel needs to be separated from other components. The ITO glass is a multi-layered surface, composed of glass panels, polarizing films, the ITO film, and the liquid–crystal panel which is glued together. Used LCDs must be dismantled to break the plastic case, remove the backlight, and retrieve the LCD panels. Manual dismantling is reported to be the most efficient and cost-effective way to recover metals compared to the mechanical way (Kopacek, 2010).

A second step would be the separation of the ITO glass from the liquid–crystal layer and the polarizing films, which are made of organic polymers such as polyethylene terephthalate, cellulose triacetate, polyvinyl alcohol, triphenyl phosphate, and benzene. Pyrolysis is a viable method of removing organic components from the glass substrate. Some studies (Lu et al., 2012; Wang & Xu, 2014) were able to remove the polarizing film and the liquid–crystal layer by heating and processing it into oils (e.g. acetic acid) and gases that can be recycled. A more environmentally friendly approach would be a combination of both physical and chemical treatments (Zhang et al., 2015). The organic layers could be softened by thermal shock (Li et al., 2009) or using acetone (Zhao et al., 2013) and then removed manually by hand. Once the ITO glass is recovered, crushing and milling

are required to reduce the size of the particles, increase surface area, and enhance the leaching process. Zheng et al. (2023) have recently reviewed the state of the art regarding the recovery approaches of indium from spent LCD screens.
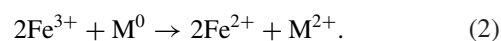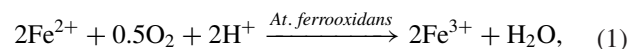
3 Bioleaching of Electronic Waste

The bioleaching of e-waste is the first step in the metal recovery process. It refers to the mobilization of base and critical metals from the solid state to their soluble form using various direct or indirect biological mechanisms. Several recent bioleaching studies and their optimal and initial conditions have been reported in Table 1.

3.1 Bioleaching Mechanisms

3.1.1 Redoxolysis

Redoxolysis is a process that involves the microbial production of compounds that can modulate the redox potential of the leaching solution. This process can occur by microbial attachment, mobilizing metals via electron transfer from the mineral to the microorganisms, but can also occur indirectly with the bioproduction of leachates oxidizing the targeted metals. The main leachate used for the bioleaching of e-waste is Fe^{3+}, which is produced by the microbially driven oxidation of Fe^{2+} (Eq. 1). Fe^{3+} can thus in turn oxidize the metals and solubilize them (Lee & Pandey, 2012) (Eq. 2). The reduced iron can then be bio-oxidized again, resulting in a cyclic process.

$$2Fe^{2+} + 0.5O_2 + 2H^+ \xrightarrow{At.\ ferrooxidans} 2Fe^{3+} + H_2O, \quad (1)$$

$$2Fe^{3+} + M^0 \rightarrow 2Fe^{2+} + M^{2+}. \quad (2)$$

Several microbes can oxidize iron, while the most commonly used microbe for mineral bioleaching is *Acidithiobacillus ferrooxidans*. It is an acidophilic chemolithoautotrophic proteobacterium that uses the energy derived from sulfur and iron oxidation for its growth (Valdès et al., 2008). Hence, the Fe^{3+}/Fe^{2+} ratio and the redox potential of the solution are key factors to monitor and understand the chemistry of the leaching solution. Among the sulfur/iron-oxidizing bacteria, *At. ferrooxidans* have been used in many recent studies involving e-waste bioleaching with very high efficiency (Arshadi & Mousavi, 2015; Becci et al., 2020; Heydarian et al., 2018; Hubau et al., 2020; Işıldar et al., 2016). Although bioleaching is mainly driven by redoxolysis, the acidic medium in which the bacterium thrives can improve the leaching efficiency.

Table 1 Literature overview of bioleaching conditions

Leaching mechanism	Microorganism(s)	E-waste	Culture medium(s)	Leaching time	Initial pH	Pulp density	T (°C)	Leaching efficiency	Reference
Redoxolysis	*Acidiphilium acidophilum*	PCB	$(NH_4)_2SO_4$ 3 g/L, $MgSO_4 \cdot 7H_2O$ 1 g/L, KH_2PO_4 0.5 g/L, KCl 0.1 g/L, $Ca(NO_3)_2$ 0.018 g/L, $FeSO_4 \cdot 7H_2O$ 0.01 g/L, S^0 25 g/L	60 d	2.5	1 g/L	30	Cu 79%, Ni 39%, Zn 29%, Pb 10%	Priya and Hait (2018)
Redoxolysis	*Acidithiobacillus ferrooxidans*	LCD	$(NH_4)_2SO_4$ 0.4 g/L, K_2HPO_4 3 g/L, $MgSO_4 \cdot 7H_2O$ 0.5 g/L, $CaCl_2 \cdot 2H_2O$ 0.25 g/L	8 d	2	15 g/L	30	In 100%	Xie et al. (2019)
Redoxolysis	*Acidithiobacillus caldus* *Leptospirillum ferriphilum* *Sulfobacillus thermosulphidooxans* *Ferroplasma*	PCB	$MgSO_4$ 0.5 g/L, $(NH_4)_2SO_4$ 0.4 g/L, K_2SO_4 0.4 g/L, $CaH_4(PO_4)_2$ 0.29 g/L	26 d	1–1.5	10 g/L	45	Cu 96.8%, Zn 68%, Al 45%	Akbari and Ahmadi (2019)
Redoxolysis	*Acidithiobacillus ferrooxidans*	LED	$(NH_4)_2SO_4$ 3 g/L, K_2HPO_4 0.5 g/L, $MgSO_4 \cdot 7H_2O$ 0.5 g/L, KCl 0.1 g/L, $Ca(NO_3)_2$ 0.01 g/L, $FeSO_4 \cdot 7H_2O$ 44.22 g/L	29 d	2–2.5	20 g/L	30	Cu 84%, Ni 96%, Ga 60%	Pourhossein and Mousavi (2018)
Redoxolysis	*Acidithiobacillus ferrooxidans*	PCB	$(NH_4)_2SO_4$ 3 g/L, K_2HPO_4 0.5 g/L, KCl 0.1 g/L, $MgSO_4 \cdot 7H_2O$ 0.5 g/L, $Ca(NO_3)_2$ 0.01 g/L, $FeSO_4 \cdot 7H_2O$ 0.23 g/L	7 d	3	36 g/L	30	Cu 87.5%	Li et al. (2015)
Redoxolysis	*Acidithiobacillus ferrooxidans*	PCB	$(NH_4)_2SO_4$ 2 g/L, K_2HPO_4 0.1 g/L, $MgSO_4 \cdot 7H_2O$ 0.25 g/L, KCl 0.1 g/L, $FeSO_4 \cdot 7H_2O$ 0.25 g/L, S^0 10 g/L	7 d	1.5	50 g/L	30	Cu>95%	Wang et al. (2018)
Redoxolysis	*Acidithiobacillus ferrooxidans* (DSMZ 1927)	LiB	$(NH_4)_2SO_4$ 0.50 g/L, KCl 0.05 g/L, $K_2HPO_4 \cdot 3H_2O$ 0.20 g/L, $MgSO_4 \cdot 7H_2O$ 0.50 g/L, $Ca(NO_3)_2$ 0.01 g/L, $FeSO_4 \cdot 7H_2O$ 45.0 g/L	3 d	2	100 g/L	30	Co 94%, Li 60%	Roy et al. (2021)

(continued)

Table 1 (continued)

Leaching mechanism	Microorganism(s)	E-waste	Culture medium(s)	Leaching time	Initial pH	Pulp density	T (°C)	Leaching efficiency	Reference
Redoxolysis	*Acidithiobacillus ferrooxidans* (DSMZ 583) *Leptospirillum ferrooxidans* (DSM 2705) *Acidithiobacillus thiooxidans* (DSMZ 9463)	PCB	$FeSO_4 \cdot 7H_2O$ 9 g/L $(NH_4)_2SO_3$ 3 g/L $MgSO_4 \cdot 7H_2O$ 0.5 g/L KH_2PO_4 0.5 g/L $Ca(NO_3)_2 \cdot 4H_2O$ 0.1 g/L S^0 1 g/L	8 d	1.8	10 g/L	30	Al 55.8% Cu 97.3% Ni 79.3% Zn 66.8%	Erust et al. (2020)
Redoxolysis Acidolysis	*Acidithiobacillus ferrooxidans* (PTCC 1647) *Acidithiobacillus thiooxidans* (PTCC 1717)	LiB	– $(NH_4)_2SO_4$ 3.0 g/L $KC1$ 0.1 g/L K_2HPO_4 0.5 g/L $MgSO_4 \cdot 7H_2O$ 0.5 g/L $Ca(NO_3)_2$ 0.01 g/L $FeSO_4 \cdot 7H_2O$ 44.2 g/L – $(NH_4)_2SO_4$ 2 g/L $K_2HPO_4 \cdot 3H_2O$ 0.25 g/L $MgSO_4 \cdot 7H_2O$ 0.25 g/L $KC1$ 0.1 g/L S^0 10 g/L	16 d	1.5	4–40 g/L	32	Li 99.2% Ni 89.4% Co 50.4%	Heydarian et al. (2018)
Redoxolysis	*Acidithiobacillus ferrooxidans* (PTCC1626)	PCB	$(NH_4)_2SO_4$ 3 g/L K_2HPO_4 0.5 g/L $MgSO_4 \cdot 7H_2O$ 0.5 g/L $KC1$ 0.1 g/L $Ca(NO_3)_2$ 0.01 g/L $FeSO_4 \cdot 7H_2O$ 44.22 g/L	20 d	1	8.5 g/L	30	Cu 100% Ni 100%	Arshadi and Mousavi (2015)
Redoxolysis Acidolysis	*Acidithiobacillus thiooxidans* (DSM 9463) *Acidithiobacillus ferrooxidans* (DSM 17,398)	PCB	$(NH_4)_2SO_4$ 2 g/L $MgSO_4 \cdot 7H_2O$ 0.25 g/L KH_2PO_4 0.1 g/L $KC1$ 0.1 g/L $FeSO_4 \cdot 7H_2O$ 8 g/L S^0 10 g/L	20 d	1	10 g/L	27	Cu 98%	Işıldar et al. (2016)
Acidolysis	*Acidithiobacillus thiooxidans* (PTCC 1717)	LCD	$(NH_4)_2SO_4$ 2 g/L K_2HPO_4 0.25 g/L $MgSO_4 \cdot 7H_2O$ 0.25 g/L $KC1$ 0.1 g/L S^0 5 g/L	15 d	2	16 g/L	30	In 100% Sr 10%	Jowkar et al. (2018)
Complexolysis	*Aspergillus fumigatus* (KUC1520) *Aspergillus flavipes* (KUC5033) *Aspergillus japonicus* (KUC5035) *Aspergillus tubingensis* (KUC5037) *Aspergillus versicolor* (KUC5201) *Aspergillus niger* (KUC5254)	Zn-Mn/Ni–Cd batteries	– ME medium: Malt extract 20 g/L – Sucrose medium: Sucrose 100 g/L $NaNO_3$ 1.5 g/L KH_2PO_4 0.5 g/L $MgSO_4 \cdot 7H_2O$ 0.025 g/L $KC1$ 0.025 g/L Yeast extract 1.6 g/L	4–8 d	2.47–7.47	1 g/L	27	Zn-Mn: Zn 100% Mn 92% Ni–Cd: Ni, Zn, Cd>90%	Kim et al. (2018)

Table 1 (continued)

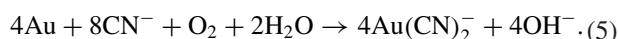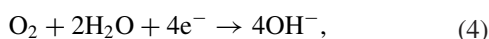| Leaching mechanism	Microorganism(s)	E-waste	Culture medium(s)	Leaching time	Initial pH	Pulp density	T (°C)	Leaching efficiency	Reference
Complexolysis	Aspergillus niger	LCD	Sucrose 50 g/L NaNO$_3$ 1.5 g/L KH$_2$PO$_4$ 0.5 g/L MgSO$_4$·7H$_2$O 0.025 g/L KCl 0.025 g/L Yeast extract 1.6/L	90 min	4	10 g/L	70	In 100%	Cui et al. (2020)
Complexolysis	Aspergillus niger	LCD	Sucrose 100 g/L NaNO$_3$ 1.5 g/L KH$_2$PO$_4$ 0.5 g/L MgSO$_4$·7H$_2$O 0.025 g/L KCl 0.025 g/L Yeast extract 1.6 g/L	15 d	2.5–3	10 g/L	30	In 100%	Cui et al. (2021)
Complexolysis	Aspergillus niger	PCB	Sucrose 100 g/L NaNO$_3$ 1.5 g/L KH$_2$PO$_4$ 0.5 g/L MgSO$_4$·7H$_2$O 0.025 g/L KCl 0.025 g/L Yeast extract 1.6 g/L	2 h	1.97	10 g/L	80	All metals 100%	Jadhav et al. (2016)
Complexolysis	Aspergillus niger (PTCC 5210)	LiB	Sucrose 100 g/L NaNO$_3$ 1.5 g/L KH$_2$PO$_4$ 0.5 g/L MgSO$_4$·7H$_2$O 0.025 g/L KCl 0.025 g/L Yeast extract 1.6 g/L	14 d	6	10 g/L	30	Cu 100% Li 95% Mn 70% Al 65% Co 45% Ni 38%	Bahaloo-Horeh et al. (2016)
Complexolysis	Aspergillus niger (PTCC 5210)	LiB	Sucrose 116.90 g/L NaNO$_3$ 1.5 g/L KH$_2$PO$_4$ 0.5 g/L MgSO$_4$·7H$_2$O 0.025 g/L KCl 0.025 g/L Yeast extract 1.6 g/L	8 d	5.44	10/20 g/L	30	Cu 100% Li 100% Mn 77% Al 75% Co 64% Ni 54%	Bahaloo-Horeh and Mousavi (2017)
Complexolysis	Chromobacterium violaceum (ATCC12471)	PCB	Peptone 10 g/L Yeast extract 5 g/L Glycine 5 g/L	7 d	3	36 g/L	30	Au 70.6%	Li et al. (2015)
Redoxolysis	Leptospirillum ferriphilum Sulfobacillus thermosulfidooxidans	PCB	(NH$_4$)$_2$SO$_4$ 3 g/L K$_2$HPO$_4$ 0.5 g/L MgSO$_4$·7H$_2$O 0.5 g/L KCl 0.1 g/L Ca(NO$_3$)$_2$ 0.01 g/L FeSO$_4$·7H$_2$O 74.5 g/L	9 d	1.5	100 g/L	55	Cu 93.4%	Wu et al. (2018)

Table 1 (continued)

Leaching mechanism	Microorganism(s)	E-waste	Culture medium(s)	Leaching time	Initial pH	Pulp density	T (°C)	Leaching efficiency	Reference
Redoxolysis	*Leptospirillum ferrooxidans Acidithiobacillus ferrooxidans Sulfobacillus thermosulfidooxidans*	PCB	$(NH_4)_2SO_4$ 0.4 g/L H_3PO_4 85% 0.81 g/L $MgSO_4 \cdot 7H_2O$ 0.52 g/L KOH 0.48 g/L $FeSO_4 \cdot 7H_2O$ 0.52 g/L	2 d	1.3	50 g/L	36	Cu 96% Zn 85% Ni 73% Co 93% Fe 93%	Hubau et al. (2020)
Redoxolysis	*Leptospirillum ferrooxidans* (DSMZ 2705T) *Acidithiobacillus ferrooxidans* (DSMZ 14882T)	PCB	DSMZ 882. *Leptospirillum* (HH) medium	7 d	1.6	50 g/L	30	Cu 91% Zn 73%	Becci et al. (2020)
Complexolysis	*Pseudomonas balearica* SAE1	PCB	LB medium 5 g/L glycine	7 d	8	10 g/L	30	Au 68.5% Ag 33.8%	Kumar et al. (2018)
Complexolysis	*Pseudomonas putida* (WSC361) *Pseudomonas fluorescens* (E11.3)	PCB	Meat extract 1 g/L Yeast extract 2 g/L Peptone 2 g/L NaCl 5 g/L Glycine	5 d	7	5 g/L	30	Au 44%	Işıldar et al. (2016)
Complexolysis	*Sphingomonas sp.* MXB8 *Candida orthopsilosis* MXL20 *Aspergillus niger* MX7, MX5, MXPE6	PCB	– Potato Dextrose Agar (DB Bioxon®)—25 mL containing 20 g Dextrose 1.5 g $(NH_4)_2SO_4$ 0.5 g KH_2PO_4 0.025 g $MgCl_2$ 0.1 g $CaCl_2$ – 25 mL containing 20 g Dextrose 1.5 g $(NH_4)_2SO_4$ 0.5 g NaH_2PO_4 0.1 g $MgCl_2$ 0.1 g $CaCl_2$	35 d	<7	–	14–24	Ag 54% Cu 2.08% Au 0.53%	Díaz-Martínez et al. (2019)
–	*Streptomyces albidoflavus* (TN10)	PCB	Yeast extract malt extract broth	3 d	6	6 g/L	28	Al 66% Ca 74% Cu 68% Cd 65% Fe 42% Ni 81% Zn 82% Ag 56% Pb 46%	Kaliyaraj et al. (2019)

3.1.2 Complexolysis

Complexolysis involves a chelating agent produced (secondary product or by-product) by a selected microorganism and metal ions to form a metal–organic complex. The chelating agent also called a ligand creates a coordinate covalent bond with the metal ions leading to their solubilization. The ligands can be organic compounds such as siderophores. These molecules are usually produced in the environment with the aim of solubilizing iron to make it available to plants and bacteria (Ahmed & Holmström, 2014). Siderophores' structure variability makes them able to chelate other metals such as In, Ga, and REE (Hofmann et al., 2020; Johnstone & Nolan, 2015).

In biohydrometallurgical processes, complexolysis is mainly carried out by cyanogenic bacteria and fungi in alkaline conditions. Cyanide (CN^-) is generally used to mobilize gold and other noble metals like Ag, Pd, or Pt. In fact, since 1898, cyanide has been widely used to extract gold from primary and secondary ores (Mudder & Botz, 2006). Gold dissolution by cyanide consists of anodic (Eq. 3) and cathodic (Eq. 4) equations and can be summarized by Eq. 5 below:

$$4Au + 8CN^- \rightarrow 4Au(CN)_2^- + 4e^-, \quad (3)$$

$$O_2 + 2H_2O + 4e^- \rightarrow 4OH^-, \quad (4)$$

$$4Au + 8CN^- + O_2 + 2H_2O \rightarrow 4Au(CN)_2^- + 4OH^-. \quad (5)$$

This reaction can be performed by many heterotrophic bacteria such as *Chromobacterium violaceum*, *Pseudomonas fluorescens*, and *Pseudomonas putida* (Işıldar et al., 2016; Li et al., 2015). Some cyanogenic bacteria can detoxify cyanide and transform it into nontoxic β-cyanoalanine (Knowles, 1976), which renders biogenic cyanide production a safer alternative than chemical cyanide treatment. Fungi such as *Aspergillus niger* have also been used for metal recovery from WEEE (Bahaloo-Horeh & Mousavi, 2017; Cui et al., 2021; Jadhav et al., 2016). They produce organic acids (e.g. malic, oxalic, citric acids) that can chelate metals and thus mobilize them.

3.1.3 Acidolysis

Acids are widely used for metal leaching in hydrometallurgy. The protons can weaken the bond between the metal ions, allowing their mobility. In case of bioleaching, the acids are produced by microorganisms, but the principle remains the same. For instance, chemolithotrophic bacteria such as *Acidithiobacillus thiooxidans* use elemental sulfur as its primary energy source and oxidize it to sulfuric acid (Eq. 6). Sulfuric acid then reacts with the metal and solubilizes it from the host material (Eq. 7).

$$2S^0 + 3O_2 + 2H_2O \rightarrow 2H_2SO_4, \quad (6)$$

$$2M^0 + O_2 + 2H_2SO_4 \rightarrow 2M^{2+} + 2SO_4^{2-} + 2H_2O. \quad (7)$$
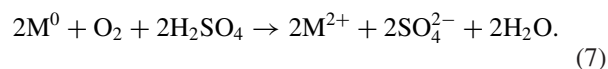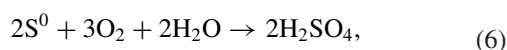
In recent bioleaching studies, acidolysis is often coupled with complexolysis and redoxolysis to enhance the leaching efficiency (Baniasadi et al., 2019; Işıldar et al., 2016). Biogenic acids are also used to control the pH of the leaching process (Heydarian et al., 2018). Still, acidolysis remains a powerful mechanism to utilize in bioleaching processes. Jowkar et al. (2018) achieved a 100% and 10% leaching rate for In and Sr, respectively, from LCD panels using only *At. thiooxidans*.

3.2 Bioleaching Optimization and Limiting Factors

3.2.1 Microorganisms

Bioleaching relies on the metabolic capacity of microorganisms to mobilize metal ions from the original substrate. The microorganisms involved produce leaching agents such as oxidizing agents (Fe^{3+}), complexing agents (CN^-), siderophores as well as organic (oxalic, malic, citric) or inorganic acids (H_2SO_4). The bioleaching efficiency lies in optimizing the production of these leaching agents. Whereas bioleaching is limited by abiotic factors such as pH, temperature, particle size, substrate concentration or nutrient, the bacterial concentration and metal tolerance are biotic factors that are critical for one-step processes (Jagannath et al., 2017; Sun et al., 2020).

Researchers have recommended the use of consortia for the leaching of e-waste. Microbial consortia can be found in a variety of habitats, where they work cooperatively to carry out metabolic activities, resulting in improved productivity over monocultures (Frey-Klett et al., 2011; Liu et al., 2011; Valix, 2017). Furthermore, the paring of suitable strains can result in reinforced metal resistance which can be attributed to physiological changes and a higher metal efflux (Ilyas et al., 2014).

3.2.2 pH

Microorganisms require optimum pH conditions for their growth and activity. Optimum pH conditions result in better leaching agents' production and bioleaching capability. Although most research on cyanogenic bacteria focuses less on optimum pH rather than initial pH, a few studies have shown that *C. violaceum* requires a slightly alkaline pH of 8 for growth, while cyanide production is maximized around pH 10 (Faraji et al., 2021; Li et al., 2015; Pourhossein et al., 2021; Shin et al., 2013). For acidophilic bioleaching involving iron-oxidizing bacteria, a pH above 2.5 causes jarosite

to precipitate and trap Fe^{3+} in a thin layer that is deposited on the waste particle surface. This results in a decreased bioleaching efficiency. Moreover, an increase in pulp density can cause an increase in pH since most waste material is alkaline in nature (Pourhossein & Mousavi, 2018; Roy et al., 2021). Thus, most research tries to maintain a pH lower than 2. Initial pH is generally adjusted with the addition of sulfuric acid and can be controlled by the production of biogenic H_2SO_4 by sulfur-oxidizing bacteria such as *At. thiooxidans* (Işıldar et al., 2016). Arshadi and Mousavi (2015) showed that the mobilization of Ni is the most impacted for PCB bioleaching, achieving the maximum recovery for Ni and Cu at pH 1. With the same substrate, Erust et al. (2020) found the optimum pH for Cu, Ni, Al, and Zn to be around 1.8. A recent study on LIB bioleaching using a combination of *At. thiooxidans* and *At. ferrooxidans* indicated the optimum pH for Co, Ni, and Li to be 1.5 (Heydarian et al., 2018).

3.2.3 Temperature

Microbial growth activity is influenced by temperature, which is reflected in bacterial bioleaching capability. Microbes are classed as psychrophiles, mesophiles (e.g. *At. ferrooxidans*), or thermophiles (e.g. *Sulfobacillus thermosulfidooxidans*) according to their temperature dependency. Extremophilic bacteria such as psychrophiles and thermophiles grow at extremely low ($-20\ °C$ to $10\ °C$) or high temperatures ($40–85\ °C$), while mesophiles thrive at temperatures ranging from $10\ °C$ to $40\ °C$ (Babu et al., 2015; Sethurajan & Gaydardzhiev, 2021). The temperature of spent medium can be increased to facilitate the dissolution of metals without changing the optimal growth temperature. This was performed by Cui et al. (2020) and Jadhav et al. (2016), where they performed the bioleaching of LCD and PCB, respectively, using *A. niger*. Temperatures of 70 °C and 80 °C were achieved, resulting in improved metal mobilization. Most bioleaching studies use mesophiles, so the optimum temperature usually lies around 30 °C. Studies that use consortia of mesophile and thermophile require temperatures around 45 °C (Akbari & Ahmadi, 2019).

3.2.4 Growth Medium

The composition of growth medium has a significant impact on the efficiency of bioleaching. A medium with an optimized substrate concentration allows the better growth of the cultivated microorganisms, as well as, improved leaching efficiency. The substrate used differs depending on the microorganism.

Chemolitotrophs such as *At. ferrooxidans* and *At. thiooxidans* utilize ferrous iron in the form of $FeSO_4$ and elemental sulfur, respectively. The primary energy source used by cyanide-producing heterotrophic bacteria is glycine and many fungi use organic substrates such as sucrose or glucose.

Since *At. ferrooxidans* is an autotrophic microorganism, the rate of iron and sulfur oxidation is partly dependent on the metabolic fixation of CO_2 (Silver, 1970; Valdés et al., 2008). Guezennec et al. (2018) performed bioleaching on a copper concentrate while increasing the partial pressure of CO_2 in the incoming gas. This study showed that the fraction of CO_2 in the air was insufficient for optimal kinetics and that enrichment along with a more active bacterial community was necessary for more efficient CO_2 uptake.

Many studies investigated the influence of Fe^{3+} concentration on bioleaching experiments. High Fe^{3+} concentration is a major constraint as it leads to the precipitation of jarosite (Eq. 8).

$$K^+ + 5OH^- + 3Fe^{3+} + SO_4^{2-} + H_2O \rightarrow KFe_3(SO_4)_2(OH)_6 + H^+. \tag{8}$$
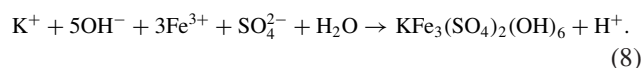
Jarosite formation decreases initial Fe^{3+} content and thus leaching efficiency. This leachate loss is a major issue for metal recovery. To overcome this issue, Wang et al. (2018) have proposed an experimental model to mobilize copper from PCBs by minimizing the initial $FeSO_4$ input and using a combination of iron-oxidizing and sulfur-oxidizing bacteria. A copper leaching rate of over 95% was achieved using only 0.25 g/L of $FeSO_4 \cdot 7H_2O$ (and 10 g/L S^0), which is 239 times lower than in similar studies.

In recent studies, statistical models are often used to determine the optimal parameters for maximum mobilization of metals in solution. In particular, the response surface method (RSM) is widely used, and its purpose is to explore the relationships between different sets of variables, whether dependent or independent. This method is usually done in central composite design (CDD), which takes the form of a polynomial quadratic equation, and allows to predict for a given set of variables the number of experiments needed to determine the desired response (e.g. percentage of metal recovered). In e-waste bioleaching experiments, these variables are typically initial pH, inoculum size, substrate concentration, pulp density, and particle size.

Following this method, Arshadi and Mousavi (2015) optimized four critical parameters for a mobile phone PCBs leaching experiment which are pH, Fe^{3+} concentration, pulp density, and particle size. It was found that Fe^{3+} concentrations between 2 and 8 g/L were suitable for better metal recovery. Erust et al. (2020) showed that a Fe^{3+} concentration of 9 g/L displayed the highest leaching yield for all metals as well as the highest ORP, compared to lower concentrations. Heydarian et al. (2018) performed the bioleaching of mobile phone LIBs using a combination of *At. ferrooxidans* and *At. thiooxidans*. For this purpose, the initial concentration of the two substrates $FeSO_4$ and S^0 were investigated. Maximum recovery was achieved with a concentration of 36.7 g/L for $FeSO_4$ and 5.0 g/L for S^0, with an initial pH of 1.5.

RSM was also used by Jowkar et al. (2018) to determine the optimum S^0 concentration for LCD bioleaching using *At. thiooxidans* and resulted in 8.6 g/L. The same experimental design was used by Bahaloo-Horeh and Mousavi (2017) to investigate the parameters for LIB bioleaching using the fungus *A. niger*, which uses organic compounds such as sucrose or glucose as substrate. They reported that the optimum sucrose concentration was 116.90 g/L. On the other hand, for the bioleaching of In from LCD panels, Cui et al. (2021) reduced the sucrose concentration from 100 to 50 g/L to increase its utilization. This resulted in an enhancement of oxalic acid production for the bioleaching of In from LCD panels. Kim et al. (2016) investigated the leaching potential of six *Aspergillus* species for the recovery of metals from Zn–Mn and Ni–Cd batteries. They were able to compare the production of organic acids using either sucrose or maltose as substrate. Citric acid production was strongly favoured by the use of sucrose medium, while maltose medium led to a higher production of oxalic acid. Greater recovery of Zn, Co, Ni, and Mn was observed using the sucrose medium. It was suggested that high concentrations of oxalic acid produce precipitates of metal complexes and thus hinder the recovery of metals (Biswas et al., 2013).

Substrate optimization was also investigated in biogenic cyanide-assisted bioleaching. Cyanogenic bacteria depend on glycine to produce cyanide. Kumar et al. (2018) report that increasing the glycine concentration from 2.5 to 7.5 g/L enhanced the metal leaching capabilities of *Pseudomonas balearica* SAE1 bacterial strain, but a higher concentration decreased the leaching of the targeted metals. This phenomenon was also observed by Işıldar et al. (2016), where a 10 g/L glycine concentration had an inhibitory effect on the cyanide production and growth of *P. fluorescens*. Li et al. (2015) reported that the quantity of cyanide produced by *C. violaceum* can be increased by adding a small amount of nutritional salts to the growth medium. There was a noticeable improvement in gold leaching efficiency with the addition of NaCl and $MgSO_4 \cdot 7H_2O$.

Hubau et al. (2018) compared the effect of two distinct culture media on the bioleaching of PCBs. For this purpose, a consortium of bacteria belonging to the genus *Leptospirillum*, *Acidithiobacillus*, and *Sulfobacillus* was cultured in continuous mode. The two media, 0 km and 0Cm, had the same composition, except for the amount of $(NH_4)_2SO_4$ which was 3.70 g/L and 0.4 g/L, respectively. Reducing the NH_4^+ content allowed a better biooxidation rate with an influent Fe^{2+} concentration of 9 g/L. The decrease in NH_4^+ concentration limited the precipitation of ammonium jarosite, thereby restricting the coating of the activated charcoal used as solid support. The author suggests that modifying the concentration of monovalent ions such as potassium or ammonium in the medium could decrease the formation of jarosite, since potassium jarosite is more stable than ammonium jarosite (Kaksonen et al., 2014). This in turn, improves the bacterial attachment onto the charcoal surface.

3.2.5 Pulp Density

Pulp density is one of the most important parameters to consider in e-waste bioleaching. It refers to the ratio of solid waste per unit of liquid media. The higher the pulp density, the greater amount of metals is available for bioleaching, which increases the potential feasibility of the processes on an industrial scale. However, most laboratory-scale studies achieve a higher metal leaching rate when working with low pulp densities. E-waste is highly concentrated in metals and toxic compounds that can be harmful to microorganisms and reduce their activity (Ilyas et al., 2010; Liang et al., 2010). Moreover, high pulp densities lead to an increase in pH and thus a decrease of protons available to react with metals (Bahaloo-horeh & Mousavi, 2017). It can also result in reduced transfer rates of oxygen and carbon dioxide, and waste particles can potentially cause mechanical damage to the microorganisms (Haghshenas et al., 2009; Roy et al., 2021).

PCB bioleaching was performed by Kumar et al. (2018) using *Chromobacterium violaceum* and *P. balearica* SAE1 at pulp densities ranging from 10 to 500 g/L. While the *Chromobacterium* strain was inhibited at 200 g/L of e-waste within 24 h, SAE1 was able to fully grow at a concentration of 500 g/L. Bacterial growth has a very important effect on the amount of leachate produced and is essential for efficient leaching. At 10 g/L waste, the amount of Ag and Au leached was 56.4% and 30.5%, respectively, while at 100 g/L, the recovery dropped to 8.3% and 1.8%. The same behaviour was observed in one of the most recent PCB bioleaching studies (Erust et al., 2020; Heydarian et al., 2018). Studies on bioleaching of LIBs show a similar effect of pulp density on metal recovery (Liu et al., 2020). Fungi bioleaching carried out by Bahaloo-horeh and Mousavi (2017) exhibits high recovery rates for low pulp density: 100% Cu, 100% Li, 77% Mn, and 75% Al at 20 g/L and 64% Co and 54% Ni at 10 g/L.

Several techniques are used to overcome the toxic effects of high pulp densities. The first is the development of an adaptation by subjecting the microorganisms to increasingly high concentrations of waste. This was done by Pouhossein and Mousavi (2018) using *At. ferrooxidans* at concentrations ranging from 5 to 25 g/L of PCB powder. The adapted bacteria culture displayed higher redox potential, bacterial concentration, and Fe^{3+} concentration than the non-adapted one at 20 g/L, which is the limit before the activity was greatly inhibited. The delay in Fe^{2+} oxidation caused by PCB toxicity can be greatly reduced by successive subcultures (Anaya-Garzon et al., 2021).

Fig. 2 Diagram illustrating the multiple-step leaching performed by Roy et al. (2021)
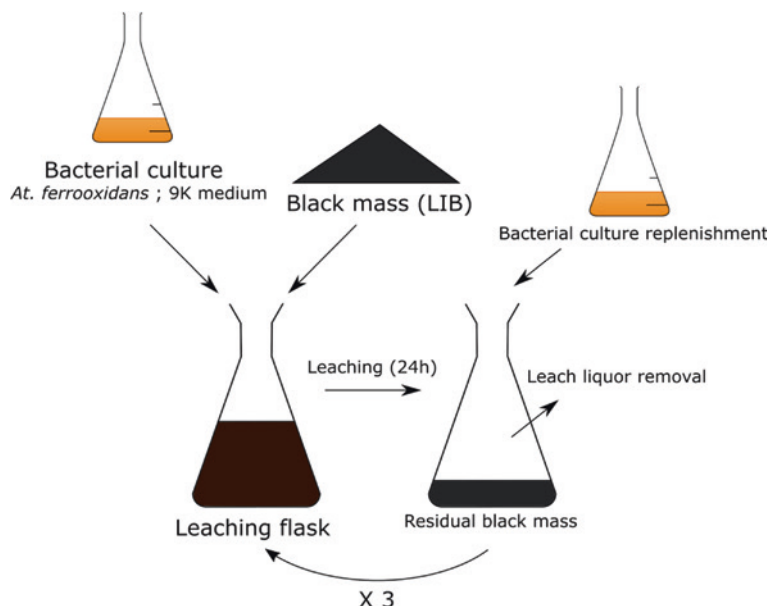

The multiple-step process has been highlighted as a successful way to diminish PCB toxicity on bacteria (Işıldar et al., 2016). Liang et al. (2010) have shown that three-step acidophilic bioleaching could improve the yield by 19% compared to a two-step approach. Moreover, it has been proven that iron-oxidative bacteria's major role is to regenerate Fe^{3+} and a non-contact mechanism is sufficient to leach copper from waste PCB (Wu et al., 2018). Fungal bioleaching of PCB has also been performed using the organic acids contained in the spent medium (Jadhav et al., 2016, Bahaloo-horeh et al., 2016). This method permits bypassing the toxicity and optimizing the production of the leachates. Roy et al. (2021) achieved a leaching rate of 94% of Co and 60% of Li at 100 g/L of LIB powder by removing the leach liquor and replenishing the bacterial culture every 24 h for a total of three cycles (Fig. 2).

## 4 Biorecovery of Critical Metal from the Pregnant Leaching Solution

Pre-treatment and bioleaching are two necessary steps in the recovery of metals from e-waste, but it is the bioprocessing of PLS that determines the economic feasibility of biotechnologies in the recovery of metals from e-waste. However, very few studies focus on the biorecovery of metals from e-waste PLS and research is at a very early stage. Promising techniques exist and have been applied successfully for the processing of PLS but mostly for the treatment of metal-rich waters such as acid mine drainage (AMD) and metallurgical leachates. A few recent biorecovery studies and key parameters are documented in Table 2.

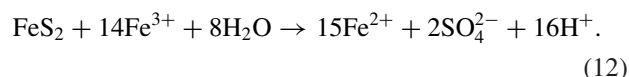### 4.1 Comparison Between AMD and E-Waste PLS and Required Pre-treatment

AMD is the result of contact between oxygen, water, and the geological subsurface during mining activities. This leads to the oxidation of minerals being in a reduced state, notably sulfides such as pyrite (FeS_2) which is the most common (Christensen et al., 1996). The oxidation of pyrite can be summarized with this reaction (Eq. 9):

$$2FeS_2 + 7O_2 + 2H_2O \rightarrow 2Fe^{2+} + 4SO_4^{2-} + 4H^+. \quad (9)$$
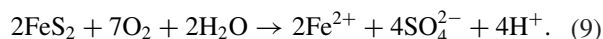
Oxidation of ferric iron is also occurring, catalyzed by iron-oxidizing bacteria such as *At. ferrooxidans* (Stumm & Morgan, 1981), following this reaction (Eq. 10):

$$Fe^{2+} + 1/4O_2 + H^+ \rightarrow Fe^{3+} + \frac{1}{2H_2O}. \quad (10)$$
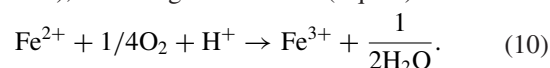
Ferrous iron then precipitates as hydroxide or jarosite at pH values above 2 (Eq. 11), and the residual Fe^{3+} participates in the oxidation of pyrite (Eq. 12) (Akcil & Koldas, 2006):

$$Fe^{3+} + 3H_2O \rightarrow Fe(OH)_3 + 3H^+, \quad (11)$$

$$FeS_2 + 14Fe^{3+} + 8H_2O \rightarrow 15Fe^{2+} + 2SO_4^{2-} + 16H^+. \quad (12)$$

The combined reaction (Eq. 13) leads to the production of acid which facilitates the dissolution of pyrite:

$$4FeS_2 + 15O_2 + 14H_2O \rightarrow 4Fe(OH)_3 + 8SO_4^{2-} + 16H^+. \quad (13)$$
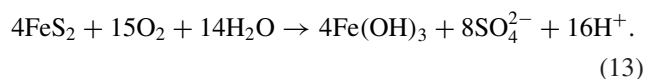
Besides the fact that other sulfides (ZnS, $CuFeS_2$) undergo similar oxidation processes, the acidic conditions of the

Table 2 Literature review of biorecovery of critical metals from pregnant leaching solutions

Mechanism	Microorganism(s)	E-waste	Culture medium	Recovery time	T (°C)	Initial pH	Metal recovered	Reference
Biosorption	*Aspergillus carbonarius* dead biomass	Synthetic solution ($K_2Cr_2O_7$)	Cornflour medium	12 h	37	2	Cr(VI) 92.43%	Lakshmi (2020)
Bioreduction Nanoparticles biosynthesis	*Bacillus licheniformis* (FZUL-63)	Synthetic polymetallic solution	LB medium	48 h	30	7	Au 90.4%	Cheng et al. (2019)
Biosorption	*Chlorella vulgaris*	Nd–Fe–B hard disk magnets	/	90 min	35	5	Nd 63.96%	Kucuker et al. (2017)
Bioreduction Nanoparticles biosynthesis	*Desulfovibrio desulfuricans* (ATCC 29577)	Electronic scrap	Post-gate medium C	Three-step process: 24 h, 24 h, 4 h	/	/	Cu>99% Pd>95%	Creamer et al. (2006)
Bioreduction Nanoparticles biosynthesis	*Magnetospirillum* MTB consortia	PCB						Sannigrahi and Suthindhiran (2019)
Bioreduction Carbonates	Metal reducing bacteria	PCB	$NaHCO_3$ 2.5 g/L $CaCl_2 \cdot 2H_2O$ 0.08 g/L NH_4Cl 1.0 g/L $MgCl_2 \cdot 6H_2O$ 0.2 g/L NaCl 10 g/L Yeast extract 0.5 g/L HEPES 7.2 g/L Trace mineral 10 mL Vitamin 1 mL	14 d	25	7.7	Mn	Yumi Kim et al. (2018)
Bioreduction Sulfides	Mixed bacterial culture (SRB)	Ni–Cd battery	KH_2PO_4, 0.5 K_2HPO_4, 0.1 $NaHCO_3$ 0.5–0.8 NH_4Cl 0.3 $CaCl_2 \cdot 2H_2O$ 0.05 $NaMo_4 \cdot 2H_2O$ 0.03 KCl 0.05 $CoCl_2 \cdot 6H_2O$ 0.02 $MgCl_2 \cdot 6H_2O$ 0.05 $NiSO_4 \cdot 6H_2O$ 0.01 Sodium ascorbate 0.05 Sodium thioglycollate 0.05	10 d	30	7		Paul et al. (2020)
Bioreduction Sulfides	SRB	Synthetic spent catalyst solution	NH_4Cl 1 g/L $Na_2SO_4 \cdot 7H_2O$ 4.15–5.5 g/L $CaCl_2 \cdot 6H_2O$ 0.06 g/L $MgSO_4 \cdot 7H_2O$ 0.06 g/L Yeast extract 0.1 g/L KH_2PO_4 0.5 g/L Ascorbic acid 0.05 g/L Nathioglycollate 0.05 g/L $FeSO_4 \cdot 7H_2O$ 0.1 g/L Sodium lactate	90 d	20–25	2	Mo 72% Co 20%	Cibati et al. (2013)
Bioreduction Sulfides	SRB	Synthetic AMD	Glycerol 5 mM Yeast extract 0.1 g/L	100 d	30	2.2	Cu	Colipai et al. (2018)
Bioreduction Sulfides	SRB	Metallurgical leach residues	K_2HPO_4 0.5 g/L NH_4Cl 1.0 g/L $CaSO_4$ 1.0 g/L $FeSO_4 \cdot 7H_2O$ 0.5 g/L Sodium lactate 3.5 g/L $MgSO_4 \cdot 7H_2O$ 2.0 g/L Yeast extract 1.0 g/L Ascorbic acid 0.1 g/L	15 d			Zn 97%	Sethurajan et al. (2017)

Table 2 (continued)

Mechanism	Microorganism(s)	E-waste	Culture medium	Recovery time	T (°C)	Initial pH	Metal recovered	Reference
Bioreduction Sulfides	SRB community	LiB	K_2HPO_4 0.5 g/L NH_4Cl 1.0 g/L Na_2SO_4 1.0 g/L $MgSO_4 \cdot 7H_2O$ 2.0 g/L $FeSO_4 \cdot 7H_2O$ 0.5 g/L $CaCl_2 \cdot 2H_2O$ 0.1 g/L Sodium thioglycolate 0.1 g/L Ascorbic acid 0.1 g/L Yeast extract 1.0 g/L Sodium-DL-lactate 2.0 g/L	181 d	30–40	7.5	Al 99% Ni 99% Co 99% Fe 99.5% Mg 49.1% Mn 98.9% Cd 98.6% Zn 98.4% Cu 99.9%	Calvert et al. (2019)

environment lead to the dissolution of other metals associated with mining activities such as cadmium, zinc, copper, lead, and arsenic (Stumm & Morgan, 1996). Thus, AMDs can be characterized as aqueous, highly acidic matrices that contain high concentrations of dissolved metal and sulfate ions in which microorganisms are key contributors to mineral leaching. In this regard, AMDs are extremely similar to acidic e-waste PLS resulting from bioleaching processes such as redoxolysis and acidolysis. Since very few studies focus on real e-waste bioleachate, AMD biotreatment methods for the recovery of metal could be a reliable source of insight for future e-waste downstream bioprocesses.

As previously mentioned, most of the bioleaching processes are taking place in acidic conditions and iron is either added in the leaching solution as oxidant (Yazici & Deveci, 2014; Sethurajan & van Hullebusch, 2019; Wang et al., 2018) being regenerated by iron-oxidizing bacteria or originating from the leaching of WEEE material processed (Işıldar, 2019). In view of the heterogeneity in metal composition of WEEE materials and selective nature of bioleaching systems, many metals dissolve in the leaching process, leading to the undesired contamination of PLS. PLS mostly originating from waste of printed circuit boards (WPCBs) leaching contains many impurity metals (Fe in particular, mostly present in its ferrous form) at significant levels (Işıldar, 2019). These impurities should be reduced to acceptable levels to avoid problems in downstream recovery processes, e.g. low current efficiencies in electrowinning of copper from sulfate solutions in the presence of ferric/ferrous ions (Wang et al., 2018) or base metal sulfide sequential precipitation (Sethurajan & van Hullebusch, 2019). For instance, the PLS can be purified by goethite (FeOOH) and jarosite ($NaFe(III)_3(OH)_6(SO_4)_2$) precipitation before being processed for recovery step (Wang et al., 2018; Yazici & Deveci, 2014). Jarosite precipitation is widely applied in hydrometallurgy to remove iron from PLS (Miettinen et al., 2019). The precipitation as jarosite also allows the

removal of sulfate ion which usually contained in the PLS as sulfuric acid is often used for performing the acidic (bio) leaching. Wang et al. (2018) showed that iron can be readily removed (in the form of goethite) from synthetic/real leach solutions of WPCBs to a large extent ($\geq 93\%$) at the expense of high copper losses particularly at pH 4.0. Such performance has been possible by implementing the biological oxidation of ferrous to ferric ion contained in the PLS by *At. ferrooxidans* leading to the formation of iron precipitates like FeOOH and Fe_3O_4 (magnetite). Hydrogen peroxide (H_2O_2) addition, before the precipitation with NaOH, allows overcoming the poor chemical oxidation of Fe^{2+} with yields higher than 85% as reported by Amato et al. (2020) allowing also the formation of iron oxides' precipitates.

4.2 Bioprecipitation

Precipitation is a rather straightforward approach to the recovery of base metals. It consists of shifting the chemical equilibrium of a system by causing a reaction in which one of the products is poorly soluble in the reaction solvent. This is most commonly achieved with the addition of precipitants such as hydroxides (NaOH, KOH, $Ca(OH)_2$), carbonates ($CaCO_3$), and sulfides (Na_2S, NaHS, H_2S) (Huang & Yang, 2021; Michalkova et al., 2013). In bioprecipitation processes, the formation of minerals is the result of an interaction between metabolic by-products and the elements in the matrix. Precipitation is dependent on the solubility of the solid in a given solvent, with PLSs generally in the form of an aqueous matrix. The solubility of metal compounds depends on several factors: pH, the concentration of ions in solution, and temperature. In particular, pH adjustment plays an important role in the selective recovery of metals. The use of bases and acids to regulate the pH of the solution is necessary to achieve the desired precipitation pH.

4.2.1 Sulfide Bioprecipitation

Precipitation and bioprecipitation are widely used techniques for the treatment of AMD. The traditional method is to use an alkali to raise the pH and precipitate the metals (Tabak et al., 2003). Bhattacharya et al. (1979, 1981) showed that the use of sulfide for the treatment of AMD displays significantly superior removal rates compared to lime and sodium hydroxide. Moreover, sulfide precipitation presents several advantages compared to hydroxide precipitation: metal sulfides have low solubility products which is practical for acidic solutions; selectivity is achieved much more efficiently; reaction kinetics are faster; less sludge is produced due to the high density of sulfides (Cibati et al., 2013). Bioprocesses based on sulfate reduction have recently emerged as promising alternatives to traditional chemical metal-containing water treatment and metal recovery. Biogenic sulfides are produced by anaerobic bacteria called "sulfate-reducing bacteria" (SRB) which play a crucial part in the carbon and sulfur cycle (Muyzer & Stams, 2008). SRB respire using organic carbon sources to reduce the sulfate ions to hydrogen sulfide (Eq. 14). Biogenic hydrogen sulfide gas then reacts with metal cations to precipitate insoluble metal sulfide (Eq. 15).

$$2CH_2O + SO_4^{2-} \rightarrow H_2S + 2HCO_3^-, \qquad (14)$$

$$H_2S + M^{2+} \rightarrow MS\,(s) + 2H^+. \qquad (15)$$
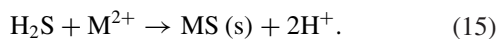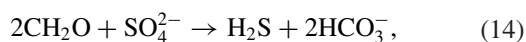
($2CH_2O$: organic carbon source; M^{2+}: metal cations).

One of the main advantages of biogenic sulfur precipitation is the selectivity of the process, which is highly dependent on pH. Sethurajan et al. (2017) studied the selective recovery of Zn from metallurgical leach residues. By removing first more than 85% of Fe through the neutralization of the bioleachate with the addition of NaOH, a removal rate of 97% of Zn was achieved. In fact, Fe is one of the major factors that affects the selective precipitation of sulfides as it can precipitate at low pH (2–3), forming jarosite or goethite (Hu et al., 2012). Calvert et al. (2019) performed a non-selective biogenic sulfide bioprecipitation of metals from a LIB leachate in a fluidised bed reactor (FBR). This study displayed excellent recovery rates (>98%) for most metals. The authors stated that selectivity could be achieved by optimizing precipitation pH and reagent concentrations. Cibati et al. (2013) achieved selectivity and high purity indexes with a less complex leachate which contained only four metals (Mo^{6+}, Ni^{2+}, Co^{2+}, V^{4+}). However, vanadium was recovered as hydroxide and the separation of Co and Ni sulfide was unfeasible due to their solubility product being very similar (Pauling, 1970). The pH of each precipitation vessel was adjusted using NaOH. Colipai et al. (2018) investigated the selective synthesis of covellite (CuS) nanoparticles from a polymetallic synthetic

AMD using acidophilic sulfate-reducing bacteria (aSRB) in a two-step process without any pH adjustment. Also, Janyasuthiwong et al. (2016) achieved 90% Cu recovery by using continuous SRB bioreactor that could precipitate Cu from PCB leachates (1.0 M HNO_3). Calvert et al. (2019) reported that over 99% precipitation efficiency was achieved for Al, Ni, Co, and Cu using biogenic sulfide from lithium-ion battery (LIB) leachates. Paul et al. (2020) reported that Cd and Ni were microbially recovered from digested Ni–Cd battery liquor as Ni and Cd sulfides' nanoparticles that have potential to be used as semi-conducting material.
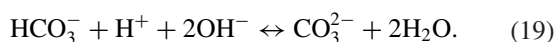
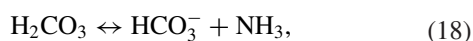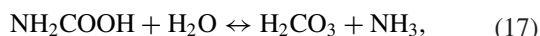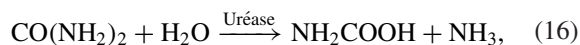### 4.2.2 Reductive Bioprecipitation

Reductive bioprecipitation describes the biologically assisted reduction of metals or metalloids (generally, precious metals) from their ionic form to a less soluble state. It occurs either through direct contact with the cell surface or via extracellular electron transfer (EET) and metal chelators (Manzella et al., 2013). Although the nature of the metal that can be reduced is entirely dependent on the microorganism and its enzymatic activity, some bacteria such as *D. desulfuricans* are active for a range of metals (Fe(III), U(IV), Cr(VI)) through the use of the hydrogenase enzyme. Similarly, many bacteria and archaea that can reduce Fe have been shown to reduce Au(III) to insoluble Au(0) (Kashefi et al., 2001). Thus, selectivity in reductive bioprecipitation processes ought to be achieved through different pathways.

The biological reduction of precious metals to metallic nanoparticles has attracted some attention in recent years (Pat-Espadas & Cervantes, 2018; Wong-Pinto et al., 2020). The production of nanomaterials from waste streams using microbes is particularly interesting because: (i) the nanomaterials can be directly reuse in catalytic/industrial processes, (ii) the microbial biomass is considered as economical catalyst and the bioconversion can be conducted at ambient temperature and pressure. The treatment of electronic waste leachate by Creamer et al. (2006) with *D. desulfuricans* is a key reference in this matter. In a three-step process, it was possible to precipitate Au^0, Pd^0, and Cu^{2+} separately. First of all, high content of Cu^{2+} inhibited the hydrogenase-driven removal of Pd^{2+} which enabled the extracellular precipitation of Au^0. Secondly, a fraction of *D. desulfuricans* biomass was pre-treated with the addition of Pd^{2+}, which accumulated on the biomass in the absence of Cu^{2+}. These "palladised cells" catalyzed the reduction of Pd^{2+} to Pd^0. The reduction of Au and Pd is enabled by the sparging of H_2. At last, the Pd-depleted leachate was treated with dimethyl disulfide produced by *Klebsiella pneumoniae* and *Escherichia coli* (Essa et al., 2006) and >99% of Cu^{2+} was recovered as a mix of sulfate and hydroxide.

Fungi are also able to accumulate Au from chloroauric acid solutions ($HAuCl_4$) both intra- and extracellularly. Au(III) reduction in Au(0) nanoparticles (AuNPs) can occur both within the cell wall but also inside cell compartments, such as the plasma membrane, the cytoplasm, and/or the nucleus (Bindschedler et al., 2017). In view of their metabolic capability, fungi should be able to first mobilize Au from e-waste through leaching processes and second to ideally immobilize it in a form that is easily recovered through biomineralization processes (AuNPs synthesis) or biosorption (Bindschedler et al., 2017).

4.2.3 Microbially Induced Carbonate Precipitation (MICP)

MICP is a biological process that uses the metabolic activity of microorganisms to precipitate carbonates. Bioprecipitation of calcium carbonate is a geochemical phenomenon that is already well known as it is common in seawater, sediments, freshwater, soil (Castro-Alonso et al., 2019) and involves several mechanisms including sulfate reduction, denitrification, ammonification, ureolysis, and photosynthesis (Zhu & Dittrich, 2016). In the last 20 years, MICP has received particular attention for its applications in solidification of biocements (Belie, 2010; Ramachandran et al., 2001), soil improvement (Ashraf et al., 2017; Wang et al., 2017) and for the remediation and immobilization of heavy metals (Achal et al., 2011; Li et al., 2013; Qiao et al., 2021). Hydrolysis of urea is the most employed metabolic pathway, generally involving the bacillus *Sporosarcina pasteurii*. Ureolysis-driven carbonate precipitation occurs in several steps: first, the urease enzyme hydrolyzes urea into ammonia and carbamic acid (Eq. 16) which spontaneously reacts with water to form ammonia and carbonic acid (Eq. 17). Ammonia is then transformed into ammonium (Eq. 18), a reaction that releases hydroxides ions which lowers the pH and induces the carbonate ions' formation (Eqs. 19 and 20).

$$CO(NH_2)_2 + H_2O \xrightarrow{\text{Uréase}} NH_2COOH + NH_3, \quad (16)$$

$$NH_2COOH + H_2O \leftrightarrow H_2CO_3 + NH_3, \quad (17)$$

$$H_2CO_3 \leftrightarrow HCO_3^- + NH_3, \quad (18)$$

$$HCO_3^- + H^+ + 2OH^- \leftrightarrow CO_3^{2-} + 2H_2O. \quad (19)$$

Finally, divalent cations (e.g. Ca^{2+} or divalent metal M^{2+}) react with carbonate ions to precipitate as carbonates (Eq. 20).

$$M^{2+} + CO_3^{2-} \leftrightarrow MCO_{3(s)}. \quad (20)$$

MICP has rarely been used for the downstream processes of e-waste leaching solutions. Rhodochrosite ($MnCO_3$)

was precipitated by Kim et al. (2018) from a waste mobile phone leaching solution using a metal reducing bacteria isolated from soil sample. Although the initial purpose of the experiment was to form reduced metals, it was suggested that $MnCO_3$ precipitation was initiated by the production of bicarbonate as a metabolic by-product.

The immobilization of heavy metals via the ureolytic pathway has shown promising results. Li et al. (2013) achieved high removal rates (88–99%) of Cd, Ni, Cu, Pb, Co, and Zn from a synthetic solution of metal chloride. The experiments were performed using a bacterium isolated from soil, as well as, *Sporosarnica pasteurii* and *Terrabacter tumescens*. The precipitates were identified as carbonates and showed rhombohedral ($NiCO_3$, $CoCO_3$), spherical ($CuCO_3$, $CdCO_3$), and needle-shaped ($PbCO_3$, $ZnCO_3$) structures.

Qiao et al. (2021) tested the heavy metal removal effect of isolated MICP strains from mining soils. The selected ureolytic consortium showed excellent performance in immobilizing Cd, Cu, Pb, Ni, and Zn, with removal rates up to 98%. The author emphasizes that the volume of inoculation must be controlled for the precipitation of carbonate crystals. The NH_3 produced during the hydrolysis of urea can lead to the formation of ammonia liquor which may have caused the dissolution of the precipitates (Stephen, 1975). Thus, except for Cd which does not seem to be affected, the optimal volume of bacteria would be between 1.25 and 4%. Only Cd and Pb precipitates could be characterized as carbonates by X-ray diffraction, while copper precipitates were identified as hydroxides. This may indicate that copper removal is mainly driven by the increase in pH during urea hydrolysis and that hydroxide and carbonate precipitation both compete for copper removal.

4.3 Biosorption

Biosorption is a physicochemical and metabolically independent biological process based on a variety of mechanisms including sorption (absorption and adsorption), ion exchange, surface complexation, and precipitation that represents a biotechnological cost-effective innovative way for the recovery of base, precious, and critical metals from aqueous solutions.

Additionally, for this extracellular process, live cells are not required; therefore, the process can be performed under extreme temperature and pH conditions (high and low, respectively). This results in an advantage from an industrial point of view as the process conditions' parameters are less difficult to manage. However, if live cells are used, greater attention needs to be paid in order to control the process and achieve the highest efficiencies. Another advantage of the extracellular process is that recovered materials

can be easily retrieved by centrifugation and filtration. The main disadvantages of this process are the low volume of metals/metalloids recovered and the long process duration.

Applying biosorption to metal recovery from e-waste is a viable option. Chemical processes including complexation, redox reactions, chelation, and precipitation take place, as well as physical interactions such as ion exchange between the biosorbent and the metals in solution. Each biosorbent has a particular set of properties depending on its functional groups which determine its biosorption capacity. The biosorbent and the operating conditions, such as pH, temperature, biosorbent concentration, and metal charge, determine the efficiency of the process.

Processes involving the use of bacteria for metal recovery from pregnant leachate solutions have been used in the hydrometallurgical industry for a long time, as well as for metal recovery from mine drainage. The practices have led the industry towards the search for environmentally friendlier and non-harmful processes. However, as it was previously mentioned, the biosorption process takes place in an aqueous phase, which poses a major setback for its application to e-waste recycling since metals have to be first leached out of the solid. In addition, in order to maximize the capacity of the biosorbent, it is necessary to chemically modify its surface. These factors make the process feasible only at laboratory scale, requiring further investigation for its scale-up and commercialization (Ramanayaka et al., 2020).

Several review papers have detailed the sorption mechanism as well as the physicochemical conditions influencing the sorption efficiency (Das & Ting, 2017; Dodson et al., 2015; Fomina & Gadd, 2014; Gadd, 2009; Gupta et al., 2019; Mack et al., 2007; Volesky, 2007; Zeraatkar et al., 2016). However, a limited number of reports are available with respect to the recovery of base, precious and critical metals from WEEE in the context of biosorption: living or inactive biomass from bacterial (Brewer et al., 2019), fungal (Sinha et al., 2018), microalgae (Čížková et al., 2019; Kucuker et al., 2017); macroalgae (e.g. *Ascophyllum nodosum*, Pennesi et al., 2019), or plant origin (e.g. persimmon tannin, Gurung et al., 2013; wood pulp, Varshney et al., 2017). Metal containing solutions such as PLS are often characterized by a pH value below 3 as well as complex composition including different competing elements that affect the biosorptive properties. Consequently, the major challenges of biosorption approaches are stability of the sorbing materials, selectivity, effectiveness, and cost efficiency.

A study of a biosorption–bioreduction process for the recovery of Te and Cd from used solar cells was published by Rajwade and Paknikar (2003) as reported in Nancharaiah et al., (2016). Using *Pseudomonas mendocina,* to remove Ag and Cd from the acid leachate of the solar cells, Te was recovered as TeO precipitate. A more recent study describes the use of *Pseudomonas* sp. strain EPR3 for

directly releasing TeO from insoluble materials including tellurium dioxide (TeO_2), cadmium telluride (CdTe), and bismuth telluride (Bi_2Te_3) (Bonificio & Clarke, 2014).

4.3.1 Bacterially Mediated Surface Adsorption

Microbially mediated surface adsorption offers a potentially cost-effective and eco-friendly approach for metal recovery. Microorganisms (bacteria and fungi) have long been exploited as biosorbents for base, precious, and critical metals' extraction, most notably for wastewater treatment. Microbial surface adsorption is a potential method for recovering metals from aqueous solutions containing metals extracted from WEEE PLS. However, there is limited research on WEEE resource recovery using this method due to the limited binding specificity of the surface functional groups, which can bind many cations with high affinity. Pollmann et al. (2018) reviewed some recent biotechnological developments in the field of biosorption and its use for critical metals' recovery from different solid waste streams including e-waste. However, only few of these developments are commercialized due to high implementation costs in comparison to conventional methods as well as its low element selectivity. Several studies have focused on the addition of metal-binding peptide tags onto bacterial surface proteins, and they have been shown to sequester more metal such as critical elements than controls. Nevertheless, these peptide tags offer limited specificity to the metals that are sorbed (Pollmann et al., 2018; Zhu et al., 2019).

In view of the increasing demand for rare earth elements (REEs) which are indispensable components of many green technologies (Dev et al., 2020; Park et al., 2020; Yu et al., 2020), numerous studies dedicated to the use of engineered bacteria have been published. Several studies have shown significant REE adsorption capacity of various microbes, highlighting their potential for REE extraction (Dev et al., 2020). For instance, Arunraj et al. (2019) reported the use of yeast (*Saccharomyces cerevisiae*) immobilized in a cellulose matrix as an efficient sorbent for the recovery of Eu(III) from an aqueous medium containing spent fluorescent lamp powder (Arunraj et al., 2019). The biosorbent regeneration was accomplished by using EDTA as complexing agent for Eu(III). The biosorbent reached a maximum adsorption capacity of 25.91 mg g^{-1} as determined by Langmuir isotherm model (Arunraj et al., 2019). However, the poor metal-binding specificity of the cell wall functional groups (e.g. carboxyl and phosphoryl) offers challenges for selective enrichment of REEs. To improve specific binding of metal ions, bioengineering approaches have been used as a cost-effective means to achieve selective metal-binding peptides and proteins on the cell surface. Lanthanide-binding tags (LBTs) made of 15–20 amino acids can complex terbium (Tb(III)) ion with high affinity (Sculimbrene & Imperiali, 2006). The Tb(III)

adsorption capacity of *E. coli* was reported to increase to 28.3 ± 1.2 mg g^{-1} dry cell weight after the expression of the fusion protein OmpA-LBT on the membrane surface, which is twice as high as that observed in the un-induced control (Park et al., 2017). By using an emulsion technique, a high cell density *E. coli* strain that had been previously modified to display lanthanide-binding tags on the cell surface was enclosed inside a permeable polyethylene glycol diacrylate (PEGDA) hydrogel. By functionalizing the OmpA protein with 16 copies of an LBT, Park et al. (2016) have previously characterized modified *E. coli* cells for improved cell surface-mediated extraction of REEs. Park et al. (2016) showed through biosorption tests using leachates from rare earth and metal-mine tailings that functionalizing the cell surface with LBT produced a number of distinguishable benefits over the non-engineered control. First, a 2–10-fold improvement in the distribution coefficients for various REEs demonstrated improved REE adsorption efficiency from all leachates. Second, with the exception of Cu, the relative affinity of the cell surface for REEs increased compared to non-REEs. Thirdly, LBT display provided a way to distinguish between valuable heavy REEs and less valuable light REEs by progressively increasing the affinity of the cell surface for REEs as a function of decreasing atomic radius. Later on, Chang et al. (2020) showed that this bioengineered *E. coli* displays an increased sorption capacity and affinity for rare earths. The engineered LBT sites have higher affinity for rare earths than wild-type surface sites and REE-cell surface binding mode depended on both pH and aqueous concentration as displayed in Fig. 3. Brewer et al. (2019) have discussed the creation of a unique flow-through biosorption-based technique for the selective recovery of REE from electronic waste (NdFeB magnet). According to Brewer et al. (2019), this microbe bead adsorbent had a uniform distribution of cells whose surface functional groups were nevertheless readily accessible and efficient for the selective adsorption of REEs. With the aid of groundbreaking research, it was shown that successful Nd extraction at flow rates of up to 3 m h^{-1} at pH 4–6 could be achieved using microbe beads packed into fixed-bed columns. After nine consecutive cycles of adsorption and desorption, the microbe bead columns were stable enough to be reused and still held 85% of their sorption capacity. When compared to non-REE impurities, REEs showed a two-bed volume increase in breakthrough points, and the adsorbed fraction of the REEs was 97% pure after breakthrough, according to a bench-scale breakthrough curve with a NdFeB magnet leachate. These findings pave the path for a biomass-based REE recovery system by showing that the microbe beads can repeatedly separate REEs from non-REE metals in a column system. The performance of incorporating a biosorption technique into a large-scale process to create commercially viable total rare earth oxides (TREOs) from diverse feedstocks was studied in Jin et al. (2017) techno-economic study of the aforementioned approach. This study established the economic viability of applying biosorption to low-grade feedstocks for REE recovery.

4.3.2 Microalgae Mediated Surface Adsorption

Among widely available biosorbents, microalgal biomass possesses relatively high binding capacities for several base, precious, and critical elements (Aksu, 2002; Monteiro et al., 2012; Wilde & Benemann, 1993). In view of their

Fig. 3 Diagram of the biosorption-based REE recovery process using crushed e-waste. Crushed e-waste was acid leached after preprocessing, such as crushing and milling, which is generally non-selectively released REEs into solution. The REEs are subsequently separated from competing metal ions using a biosorption/desorption process once the pH of the REE-bearing leachate solutions has been adjusted. This procedure results in concentrated mixed rare earth solutions. The remarkable stability of LBT even after going through numerous adsorption/desorption cycles supports the reuse of biosorbents (adapted from Park et al., 2020)
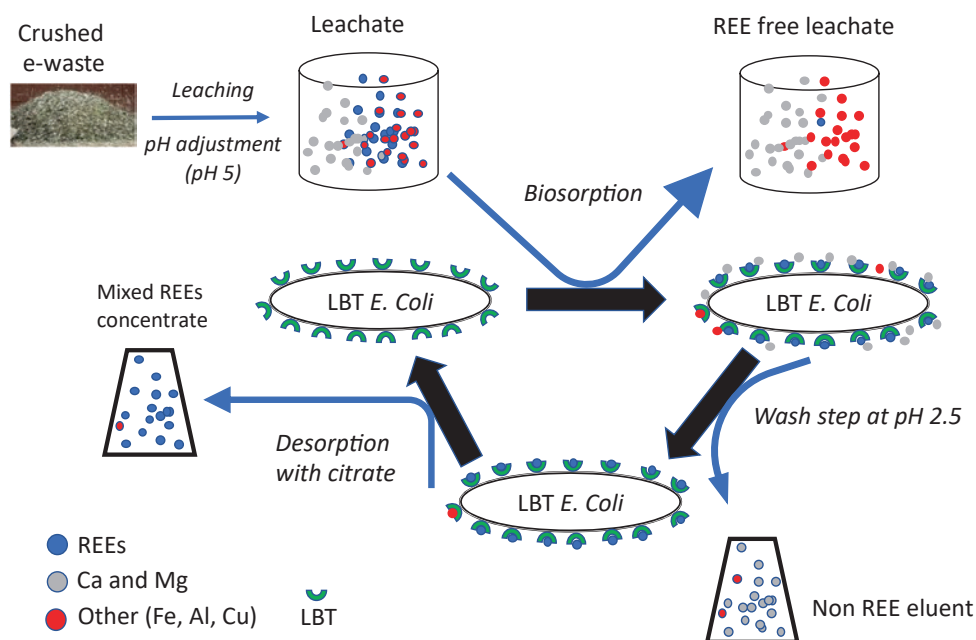
inexpensive growth requirements (solar light and CO_2) and the advantage of being utilized simultaneously for multiple technologies (e.g. carbon mitigation, biofuel production, and wastewater treatment), microalgae are also seen as suitable candidates for eco-friendly biosorption technologies (Kumar et al., 2018). Microalgae generally display significant surface area-to-volume ratio available for contact with the surrounding environment (Monteiro et al., 2012), and this property provides an additional advantage for biosorption leading to the recovery of critical metals. This is explained by the presence of carboxylic, hydroxyl, amino, phosphate, and sulfhydryl functional groups in the microalgal cell wall that can act as metal-binding sites (Kumar et al., 2018). Microalgae have been used for the recovery of several critical elements from different wastewater streams; however, only a few have been applied so far for PLS. For instance, Kücüker et al. (2017) used dried green microalgae (*Chlorella vulgaris*) to study Nd removal from neodymium permanent magnet leachates in batch and continuous sorption systems. With a biosorbent dosage of 500 mg L^{-1} and an initial neodymium content in the mixed leachate solution of 250 mg L^{-1} at 35 °C, the maximum Nd absorption ($q = 157.21$ mg g^{-1} sorbent) was ascertained at pH 5 under these conditions. The sorption capabilities of microalgae have been reported for other critical elements such as indium and rare earth elements. Nicomel et al. (2020) investigated the In(III) adsorption by microalgal biomass in batch experiments. Adsorption isotherms have well-fitted with the Freundlich model. The estimated maximum In(III) adsorption capacity of microalgae was 0.14 mmol g^{-1}, which is higher than that of some chemically modified adsorbents reported in the literature (Nicomel et al., 2020). Selectivity for In(III) over other metals, such as Cu(II), Zn(II), and Al(III), was also observed. Furthermore, repeated cycles were performed, where the microalgae biosorbent was regenerated using 0.1 M HCl solution, along with up to 80% In(III) recovery. These results indicated the potential of microalgae for In(III) biosorption from aqueous solution. Čížková et al. (2019) investigated the growth of green microalgae on red mud and the intracellular accumulation of lanthanides. It was observed that *Desmodesmus quadricauda* was able to accumulate lanthanides to the highest level (27.3 mg/kg/day), in comparison to *Chlamydomonas reinhardtii* and *Parachlorella kessleri* (24.5 and 12.5 mg kg^{-1} per day, respectively). The accumulated lanthanide content was reported to increase in the *Desmodesmus quadricauda* within 2 days from zero in red-mud free culture to 12.4, 39.0, and 54.5 mg kg^{-1} of dry mass at red-mud concentrations of 0.03, 0.05, and 0.1%, respectively. Thus, green algae are prospective vehicles for biomining or bioleaching of lanthanides from red mud (Čížková et al., 2019). In general, if microalgae could be considered as effective biosorbent, substantial

pH adaptation may be needed prior to biosorption, which may be a bottleneck especially when the aqueous solutions contain many different elements that may potentially lead to the loss of critical elements during pH correction or strongly act a competitor during critical elements' sorption. Due to the above-mentioned bottlenecks, most biosorption processes are still at the laboratory scale and poorly applied to WEEE leachates.

Several studies have been carried out for the development of different biosorbents, not only their structure but also their metabolic pathway. Even though these studies were mainly focused on removing toxic elements from wastewaters, the developed biotechnologies can also be applied to recycling processes including e-waste (Pollmann et al., 2018).

Despite these shortcomings, biological recovery of critical metals from several End-of-Life (EoL) products through a process known as "bioleaching and biosorption" has been reported by Kucuker and Kuchta (2018) and has attractive advantages compared to conventional metal processing technologies. According to a literature survey performed by Kucuker and Kuchta (2018), a schematic diagram for the recovery of critical metals from WEEE has been proposed (Fig. 4). The process includes physical separation (pre-treatment) and bio/hydrometallurgical (bioleaching and biosorption with microalgae) process for the recovery of critical metals from WEEE under a biomining concept.

4.4   Bioelectrochemical Metal Recovery Systems

Bioelectrochemical systems (BESs) have been utilized for metal removal and recovery from different waste streams including metallurgical, industrial, and wastewaters. BES consists of an anode and a cathode chamber often separated by an ion-selective membrane (Fig. 5). In the anode chamber microbial oxidation of biodegradable material (e.g. organic carbon) releases electrons to the electrode and protons to the electrolyte. The electrons flow through a circuit to the cathode and are used to reduce metallic ions in the cathode chamber. The circuit is completed by the migration of protons from the anode to the cathode through the membrane. Depending on the relative magnitude of the redox potentials at the anode and cathode, particular metals will be precipitated at the cathode (Nancharaiah et al., 2016).

The majority of studies have used synthetic solutions and have provided proof-of-principle for a few precious and rare metal(loid)s that are frequently recovered in WEEE PLS, including Ag, Au, Co, and Se (Dominguez-Benetton et al., 2018; Guo & Kim, 2020; Nancharaiah et al., 2015, 2016; Wang & Ren, 2014). Rodenas Motos et al. (2015) could achieve high recovery rates for soluble Cu(II) as Cu(0)

Fig. 4 Process flow sheet proposed for the recycling system linking bio/hydrometallurgical and biosorption process for recovery of REEs and precious metals from e-waste (modified from Kucuker & Kuchta, 2018)
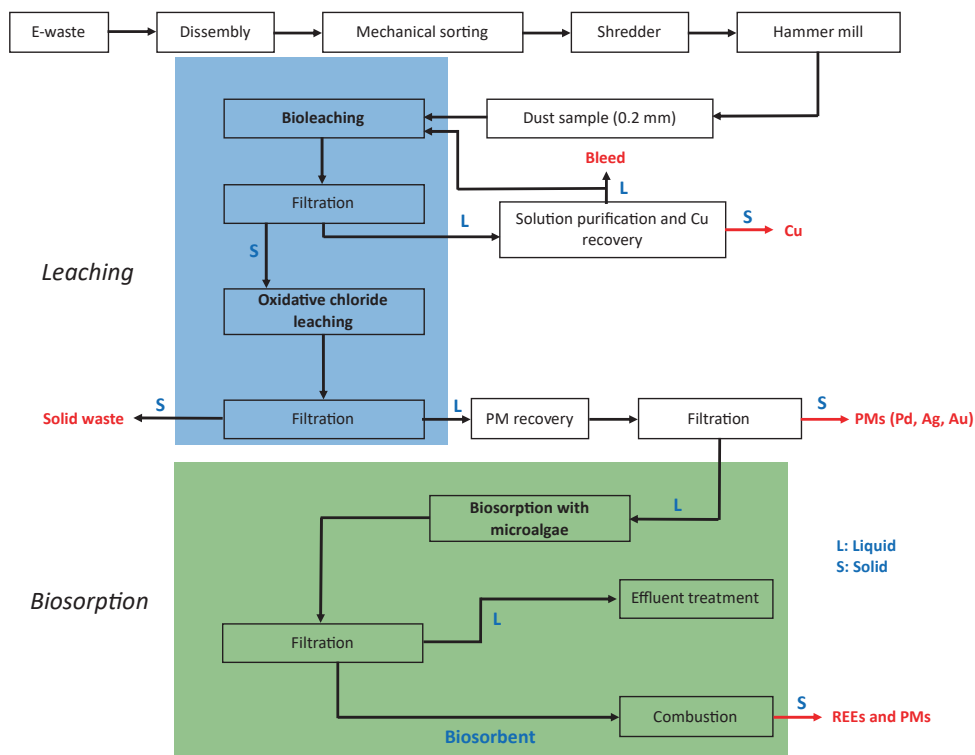
Fig. 5 Working principles of dual-chamber BES applied for metals' biorecovery. In the anode chamber, electroactive bacteria oxidize COD-rich wastewater and release reducing equivalents (electrons and protons). The electrons are used for reduction of metal ions at the cathode. Recovery of metal ions from WEEE PLS is studied through selective reduction of metal ions at the cathode. Abbreviations: *COD* chemical oxygen demand; *Me* metal; *PEM* proton exchange membrane (modified from Nancharaiah et al., 2015, 2016)
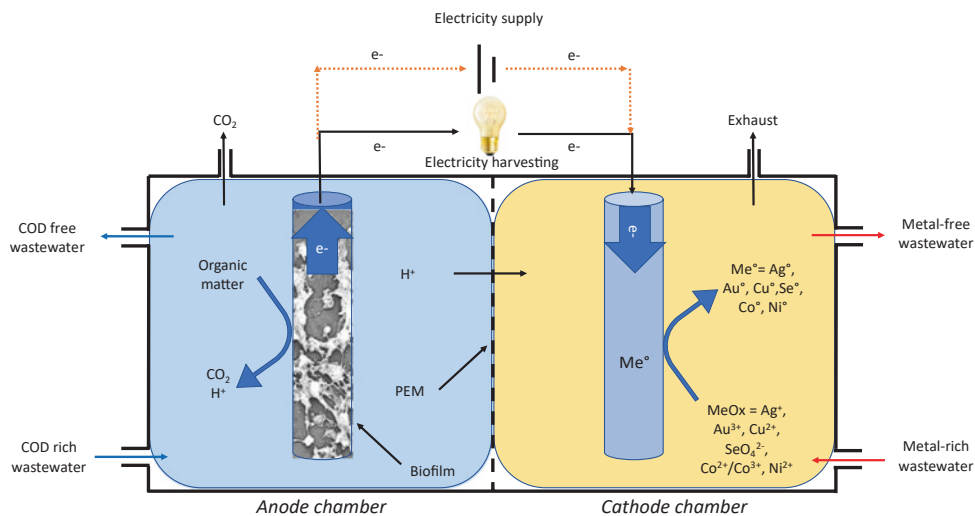
deposits. More than 90% removal efficiency was achieved in acidic solutions containing up to 2000 mg L^{-1} Cu(II). Numerous applications addressing the recovery of critical elements that are frequently found in WEEE PLS have been published. For instance, the bioelectrochemical recovery of Au^{3+}, Co^{2+}, and Fe^{3+} metal ions has been investigated by Varia et al. (2013) and the influence of aqueous gold speciation has been investigated by Ho and Babel (2019). Ho and Babel (2019) looked at the recovery of Ag and electricity production from different Ag(I)-containing synthetic wastewaters. This study found that different types of

Ag(I)-containing solutions (i.e. Ag$^+$ solution, [Ag(NH$_3$)$_2$]$^+$, [Ag(S$_2$O$_3$)$_2$]$^{3-}$ complex, and mixed multi-metal solution containing Ag$^+$, Fe^{3+}, Cu^{2+}) employed in BES reactors resulted in different performances for Ag recovery and simultaneous electricity production. In all four reactors, Ag(I) was reduced electrochemically to form deposits on the cathode surface. The highest reduction rate (i.e. kinetic reaction constant, k of 1.55 h^{-1}) was found with Ag$^+$ solution, as compared to the other Ag(I) solutions (i.e. [Ag(NH$_3$)$_2$]$^+$ complex, k of 0.117 h^{-1}; [Ag(S$_2$O$_3$)$_2$]$^{3-}$ complex, k of 0.041 h^{-1}; and mixed multi-metal solution, k of 0.129 h^{-1}).

This study shows how the aqueous solution composition may affect the removal kinetic and efficiency. Zhang et al. (2020) investigated stacked bioelectrochemical systems (BESs), composed of microbial electrolysis cells (MECs) driven by microbial fuel cells (MFCs) which provide an alternative approach for the recovery and separation of mixed W(VI) and Mo(VI) without input of external energy. In another study, Huang et al. (2019) demonstrated the feasibility of larger-scale single-chamber MECs (40 L cylindrical single-chamber MEC fed acetate) for efficient treatment of W(VI) and Mo(VI), moving metallurgical MECs closer to commercialization for PLS treatment of these two metals. All the above-mentioned studies refer to critical elements that one can find in WEEE PLS. Dominguez-Benetton et al. (2018), in a recent paper, highlighted the challenges and opportunities to turn microbial electrometallurgy into a sustainable industrial technology in the near future. Figure 6 summarizes metals studied to date using BES technology and highlights those which are highly profitable for recovery (Dominguez-Benetton et al., 2018), especially for critical metals such as PGM and lanthanides (CRM Report, 2017).

To scale up the process and study the prospect of recovering metals from WEEE leachate streams using BES, more research is still required. In order to increase the effectiveness of bioelectrochemical processes, additional research is required to understand the interactions between microbes and metals, electron transport pathways, and electrode materials (Dominguez-Benetton et al., 2018). It is still possible to further explore the BES platform's potential use in the recovery of a variety of other priceless and technologically important metals, including metals found in WEEE.

5 Integration of Bioleaching for Biorecovery Processes

5.1 The Importance of PLS Characterization

The optimal conditions for a transition from a bioleaching process to a biorecovery process are an area that needs to be addressed. It should be considered as a crucial link to complete the biohydrometallurgical loop and to strengthen the overall technology readiness. For instance, thorough characterization of pregnant leaching solutions is lacking and should be further investigated, as the application of the appropriate biorecovery method is necessary. Most PLS is not only enriched in metals from the base substrate but also comprises ions contained in the culture growth medium or resulting from metabolic production during bioleaching. It has already been mentioned previously that the heterogeneous nature of WEEE and the non-selectivity of most bioleaching processes can lead to contamination of PLS. These contaminations take the form of solvents, organic compounds, as well as, unwanted toxic metals. Depending on the method used, this chemical complexity may represent a constraint in the processing of PLS (Zhuang et al., 2015). Iron, whether employed as a nutrient for *At. ferrooxidans* bacterium or contained in different types of WEEE, is an impurity commonly found in PLS, but the presence of other elements can also inhibit the selective biorecovery of metals. Chemical reactions such as coprecipitation or sorption are commonly occurring in biorecovery studies from e-waste. Thus, additional steps between bioleaching and biorecovery might be necessary to ensure the selective precipitation of the desired metal (i.e. Kumar et al., 2022). As PLS often has extreme pH values and ionic strength, it is important to define which processes favour the existence of precipitating agents in the given pH range. For instance, biogenic carbonate ions (CO_3^{2-}) required for the precipitation of metal carbonates prevail at pH above 12.3. Thus, it is extremely challenging to integrate acidic PLS with biorecovery process based on alkaline pH. Most biorecovery studies rely on synthetic solutions containing the target metals. The solutions are prepared from metallic salts, such as metal chloride and sulfate. The resulting pH of the solution ranges from neutral to slightly acidic which is often not representative of most PLS. It is therefore essential to consider the constraints of integrating real PLS into biorecovery processes in the scope of the future studies. This would provide useful information on the actual biorecovery yields, ways to mitigate these constraints during upstream processing or bioleaching, and thus close the biohydrometallurgical loop through the best possible method.

Fig. 6 Reported metals recovered using BES. Modified from Dominguez-Benetton et al. (2018)

5.2 Selective Biomineralization and Metal Speciation

Living organisms produce mineral crystals generally as part of a process for overcoming metal toxicity. A number of innovative critical metal biomineralization processes have been studied and reported as its application has potential for the production of nanomaterials; however, mechanisms need to be further investigated as they are not completely understood (Yu et al., 2020). Particularly in e-waste recycling, biomineralization is rising as a viable technology for the recovery of platinum group metals (PGMs). Furthermore, it is a selective PGMs recovery technique which offers advantages over conventional metal recovery methods in case of complex leachates such as e-waste (Rene et al., 2017). Some case studies have shown that the recovery yield of some critical elements is strongly affected by the chemical speciation in the bulk solution. However, this aspect is too often neglected and a better knowledge of the speciation of metals in solution could help to better understand the differences in performance observed in the literature. The speciation of metallic ions in solution directly influences the sorption and reduction capacity of microorganisms. In fact, metabolic pathways such as intracellular sequestration are controlled by the speciation of metals in the medium (Worms et al., 2006). Moreover, efflux mechanisms developed by some microorganisms may change the speciation of metals by expulsing uncharged metallic atoms (Worms et al., 2006). The contribution of exopolymeric substances (EPS) to changes in metal speciation in biomineralization processes is often ignored (Gadd & Pan, 2016). Maes et al. (2016) investigated the effect of varying high salt concentrations on the recovery of platinum from a halophilic microbial community. It has been reported that high salt concentration and chloride-rich matrices may influence Pt speciation thermodynamics. In addition, a study performed in 2017 by Maes et al. used five different platinum complexes (K_2PtCl_4, K_2PtCl_6, $Pt(NH_3)_4 \cdot 2HCO_3$, cisplatin, and carboplatin) to investigate the effect of Pt speciation on platinum recovery by axenic cultures (*S. oneidensis*, *C. metallidurans*, *B. toyonensis*, and *P. stutzeri*). Not only the kinetics of the platinum bioreduction were in general faster for $Pt(II)Cl_4{}^{2-}$, but each species cultured displayed different biomineralization behaviours depending on Pt speciation and Pt complexes. The recovery of europium via anaerobic reduction by a *Clostridium* bacterial strain has been performed by Maleke et al. (2019) and showed some potential for the selective reduction of Eu^{3+} to Eu^{2+}. Such bioreduction processes involve the transition of metal from an oxidized state to a reduced state and are therefore entirely controlled by the speciation of that metal. Electrochemical bioreduction (i.e. Ho & Babel, 2019) also has the same requirements for the respective oxidation state of the target metals. To tackle and address metal speciation influence on the recovery yield, a more selective bioleaching of target metals should be further pursued, primarily through the use of organic acids and siderophores for selective extraction of targeted metals.

6 Conclusions

Biohydrometallurgy paves the way for less energy-intensive, less costly, and more environmentally friendly methods of recycling electronic waste. Although research is still in its infancy, many building blocks have been laid through the optimization of several process units. The constraints of traditional bioleaching methods and their optimal parameters are well documented, but there are still several gap areas regarding selective forms of bioleaching such as those using organic acids or biogenic siderophores. Techniques have been developed to establish a balance between ideal growth parameters and increased pulp density, which would allow industrial applicability. In order to demonstrate the industrial applicability of a fully operational biotechnological recycling process, it is essential to integrate real pregnant leaching solutions with the downstream process studies. To do so, these solutions ought to be further characterized to facilitate this transition. Overall, although bioleaching and biorecovery strategies have proven to be effective at the laboratory scale, a better understanding of biorecovery mechanisms and associated constraints will allow a higher level of technological readiness.

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