

Review



A Review on Chemical versus Microbial Leaching of Electronic Wastes with Emphasis on Base Metals Dissolution

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Abstract: There is a growing interest in electronic wastes (e-wastes) recycling for metal recovery because the fast depletion of worldwide reserves for primary resources is gradually becoming a matter of concern. E-wastes contain metals with a concentration higher than that present in the primary ores, which renders them as an apt resource for metal recovery. Owing to such aspects, research is progressing well to address several issues related to e-waste recycling for metal recovery through both chemical and biological routes. Base metals, for example, Cu, Ni, Zn, Al, etc., can be easily leached out through the typical chemical (with higher kinetics) and microbial (with eco-friendly benefits) routes under ambient temperature conditions in contrast to other metals. This feature makes them the most suitable candidates to be targeted primarily for metal leaching from these waste streams. Hence, the current piece of review aims at providing updated information pertinent to e-waste recycling through chemical and microbial treatment methods. Individual process routes are compared and reviewed with focus on non-ferrous metal leaching (with particular emphasis on base metals dissolution) from some selected e-waste streams. Future outlooks are discussed on the suitability of these two important extractive metallurgical routes for e-waste recycling at a scale-up level along with concluding remarks.

Keywords: electronic waste; recycling; chemical leaching; bioleaching; non-ferrous metal dissolution; waste printed circuit boards; spent batteries; LCD panels; waste solar panels

1. Introduction

The concomitant increment in metal demand and paucity of primary resources is an emerging challenge for valuable metal production [1,2]. In this regard, urban mining aimed towards green recycling and recovery of metals from electronic waste (e-waste) has been gaining momentum [3,4]. Additionally, the metal recycling approaches are also receiving crucial attention from the United Nations Environment Program (UNEP), since the worldwide demand for metals has been mounting consistently [5,6]. The electrical and electronic equipment (EEE) such as cell phones, smartphones, tablets, laptops, flat panel TVs, video recorders, refrigerators, digital cameras, etc., have turned out to be key requisites for the modern world [7].

E-waste or the waste electrical and electronic equipment (WEEE) consists of several parts including sub-assemblies and consumables that remain a part of the product at the time of disposal [8]. It is developing into a global concern because of its huge volume along with the health impacts pertinent to its inappropriate management and reuse. At the same time, it is also a rich source of valuable materials, which can be extracted from it judiciously for several applications [9–11]. In this regard, sustainable recycling processes would assist in increasing the metal production, while at the same time, addressing the environmental and health problems associated with hazardous wastes [5].



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). According to reports, WEEE is considered as one of the major sources of waste globally and has a high annual growth rate [12–14]. The projected worldwide e-waste generation amounted to 53.6 million ton (Mt) in 2019, which increased from 41.8 Mt in 2014 and 44.7 Mt in 2016. It is expected to grow further to 74.7 Mt by 2030 following a yearly growth rate of nearly 2 Mt [8,15,16]. Therefore, proper recycling of this waste for recovery of critical metals would be beneficial considering both from an economic and eco-friendly viewpoint [7,17,18]. According to the European WEEE Directives 2002/96/EC and 2012/19/EU, the e-waste is categorized into distinct varieties, which comprise of big and small domestic appliances, information technology and telecom equipment, monitors and control units, electrical and electronic non-industrial tools, relaxation and sports equipment, lighting devices, consumer equipment, medical non-infected devices and automatic dispensers [19].

The recycling of metals from e-wastes in a resourceful and environmentally feasible manner would assist in meeting the inadequacy of several valuable metals that have applications in technological development [20,21]. In this aspect, hydrometallurgical recycling techniques, e.g., the leaching of metals using chemicals and/or biological (re)agents have gained rapid momentum over the years [22,23]. Since, the concentration of base (nonferrous) metals, in general, is higher in e-wastes (even higher than that present in the ores); hence, this piece of review emphasizes primarily on their dissolution aspects from some selected e-waste materials/streams. Precious metals (e.g., Au and Ag), critical and/or rare metals (e.g., Li, In, Ga), also considered as non-ferrous metals [24,25], are seen to be extracted along with the base metals. In this review, very limited focus is given on the dissolution aspects of such non-ferrous metals; however, they are discussed in order to reflect the overall research attempts made with respect to the selected e-wastes. The metal dissolution aspects are reviewed in context to providing comparative insights into both the chemical and microbial leaching routes in practice. The article discusses some of the recent attempts in the area where a majority of the literature covered are within the time frame from 2015–2020, also including articles that are published in the current year 2021. To our knowledge, very limited review articles on solar panels and by-products of e-wastes are available. In this piece of review, along with some highlighted e-wastes (e.g., PCBs, batteries, LCD/LED panels), aspects related to the bio and chemical leaching of spent solar panels and certain by-products of e-wastes (e.g., flue dust, slags etc.) have been discussed.

With respect to the collection of literature, search engines such as Google Scholar, Web of Science and Scopus databases were primarily searched. Several key words such as "Electronic waste", "E-waste", "Base metal leaching from e-waste", "Non-ferrous metals", "Non-ferrous metals in E-waste", "Chemical leaching of E-waste", "Bioleaching of E-waste", "Metal leaching from spent solar panels", "by-products from e-wastes", "PCBs", "LCD panels", "Alkaline batteries", "Lithium ion batteries", "Acidophilic bioleaching of E-waste", "Fungal bioleaching", "Heterotrophic bioleaching", "Authotrophic bioleaching", "E-waste circular economy", etc., were used.

2. Hydro-Metallurgical Applications in Metal Recovery

2.1. Chemical Leaching

As part of extractive metallurgy, hydrometallurgy is a well-established process which is applied successfully for metal leaching from both primary and secondary resources [22,26]. Nowadays, several researches are being undertaken on chemical leaching of critical metals including REE [26–29], base metals [30–33] and precious metals [34–36] from e-waste materials. The process is mainly divided into three steps (leaching, solid liquid separation/purification and metal recovery). The first step consists of deriving the metal value in aqueous phase. In the second stage, the solid part is separated from leach liquor, subsequently followed by pregnant leach solution purification. The last step consists of extracting metal from the purified solution [37].

Different groups of lixiviants such as inorganic acid [30,31,33,38], organic acid [27,36] and alkaline reagents [36,39–41], are used for metal leaching from e-waste. Recently, several

reports were made with respect to REE and precious metals [26,37,42–44]. Table 1 shows a list of chemical leaching approaches that were carried out for various e-waste samples along with their experimental conditions and metal recovery. It is to be noted that the table presents base and some other non-ferrous metals, e.g., Au, Ag, Li, and In, to reflect the overall study. The following sections discuss metal leaching aspects from some selected e-waste streams with primary focus on base metals.

2.1.1. Metal Leaching from Waste Printed Circuit Boards (WPCBs)

Printed circuit boards contain a variety of base metals such as Cu, Fe, Ni, Al, Sn, Pb, Zn and precious metals including Au, Pd, and Ag. The metal composition in PCBs can vary based on the grades (high or low quality); nevertheless, Cu is the element found in a higher concentration ranging from 16 to 38.8% [45]. As per a study carried out by [46], the presence of 24.178% Cu was confirmed by the characterization of PCBs from a mobile phone. Considering the Cu content in the current primary resources (which is approximately 0.62%), the above mentioned values hold a primary importance in extractive metallurgy [47].

Table 1. List of some chemical leaching studies for metal recovery from selected e-waste streams along with their operation conditions (note: Au, Ag, Li, In, etc., have been presented, wherever applicable, to reflect the overall study along with base metal dissolution).

E-Waste	Chemical Concentration	Pulp Density	Temp	Stirring Rate	Leach Time	Metal Recovery	References
	0.5 mol/dm ³ HCl and 0.074 mol/dm ³ FeCl ₃	1/10 S/L (w/v)	Room temp.	600 rpm	24 h	96% Cu, 81% Sb	[31]
	$\frac{4 \text{ g CS}(\text{NH}_2)_2 + 2.6 \text{ g}}{\text{Fe}_2(\text{SO}_4)_3 + 3.6 \text{ N H}_2\text{SO}_4}$	1/100 S/L (w/v)	20 °C	150 rpm	7 h	100% Cu, 100% Au, 100% Ag	[48]
	1.17 M NaBr + 0.77 M Br ₂ + 2M HCl	50 g/L	23.5 °C	400 rpm	10 h	95.21% Ni, 97.88% Cu, 92.50% Zn, 97.61% Pb, 96.79% Sn, 96.52% Ag, 95.59% Au	[32]
PCBs	100 mM Fe ₂ (SO ₄) ₃	10 g/L	20 °C	300 rpm	4 h	98% Cu	[44]
	20 g/L Fe ₂ (SO ₄) ₃	1%	25 °C	200 rpm	48 h	84.3% Cu, 98.4% Ni, 100% Zn, 100% Al	[49]
	1 mol/L glycine + 10% H ₂ O ₂	1/100 S/L ratio	30 °C	400 rpm	8 h	94.08% Cu	[36]
	0.074 mol/L FeCl ₃ + 0.5 mol/L HCl	1/10 S/L ratio	Room temp.	600 rpm	24 h	96% Cu, 81% Sb	[31]
	$\frac{3.6 \text{ mol/L } \text{H}_2 \text{SO}_4 + 6\% v/v}{\text{H}_2 \text{O}_2}$	75 g/L	20 °C	-	186 min	96% Cu	[33]
	0.5 M glycine	2%	23 °C	100 rpm	72 h	96.5% Cu, 92.5% Zn, 46.8% Pb	[50]
PCBs	$0.84 \text{ M} \text{H}_2\text{SO}_4$	L/S ratio of 100:1	60 °C	200 rpm	80 min	96% Cu	[51]
Siudge	0.2 M H ₂ SO ₄	4%	25 °C	250 rpm	1 h	95% Cu	[52]
PCB dust	2 M NH ₄ OH + 17.5 M H ₂ O ₂	1%	40 °C	400 rpm	3 h	92% Cu, 50% Zn	[39]

E-Waste	Chemical Concentration	Pulp Density	Temp	Stirring Rate	Leach Time	Metal Recovery	References
	2 M H ₂ SO ₄	0.1 kg/L	80 °C	-	10 min	85–90% In	[53]
	6 M HCl	1.9 to 33.3 L/kg	-	-	2 h	968.5 mg/kg In	[54]
	1 M citrate + $0.2 \text{ M} \text{ N}_2 \text{H}_4$	20 g/L	25 °C	450 rpm	16.6 h	98.9% In	[27]
LCD -	5 M HCl	500 g/L	75 °C	400 rpm	2 h	$\begin{array}{c} 10.24 \times 10^{-3} \\ g/LSn, \\ 76.16 \times 10^{-3} \\ g/LIn \end{array}$	[55]
	1 mol/L H ₂ SO ₄	1/8 S/L ratio	70 °C	320 rpm	1 h	97.07% Sn, 9.25% In	[56]
	0.4 N H ₂ SO ₄	50%	70 °C	250 rpm	30 min	99.5% In	[57]
	0.5 M H ₂ SO ₄	0.1 g/mL	90 °C	360 rpm	2 h	98% In	[58]
	3M H ₂ SO ₄	6/1 L/S ratio	85 °C	600 rpm	10 min	76.1% Sn, 86.3% In	[59]
	2.75 mol/L H ₃ PO ₄	6 mL/g L/S ratio	40 °C	450 rpm	10 min	96.3% Mn, 99.1% Li	[60]
LIBs	1.5 mol/L malonic acid + 0.5% H ₂ O ₂	20 g/LS/L ratio	70 °C	300 rpm	20 min	98.27% Ni, 98.6% Co, 98.54% Mn, 95.74% Li	[61]
	2 mol/L H ₂ SO ₄	5 mL/g L/S ratio	60 °C	-	2 h	98% Ni, 90% Mn, 97% Co	[38]
Alkaline	H_2SO_4	10/1 L/S ratio	60 °C	300 rpm	2 h	99.2% Zn, 37.6% Mn	[62]
batteries	1.2 M glycine	10%	25 °C	210 rpm	150 min	86% Cd	[63]
	5M HNO ₃	1/35 S/L ratio	70 °C	-	180 min	96.5% Cd	[64]
	3M NaOH	L/S ratio = 50	70 °C	-	3 h	99.9% Al	[65]
-	6M H ₂ SO ₄	1/10 S/L ratio	93.2 °C	-	148 min	95.2% In	[66]

Table 1. Cont.

Hydrometallurgy has been successfully applied for the leaching of various metals. A recent study indicated that using ferric chloride (0.074 mol dm⁻³ FeCl₃) in the presence of hydrochloric acid (0.5 mol dm⁻³ HCl) led to the successful leaching of antimony (Sb) as a by-product during Cu leaching from PCBs [31]. The leaching step was followed by an intermediate precipitation process for Sb recovery (81% pure Sb) and ended with the electro-winning process for Cu recovery (96% pure copper) (Figure 1a). FeCl₃ and HCl are suitable and efficient reagents for antimony leaching [67]. Alongside these mixtures, HCl and hydrogen peroxide (H₂O₂) were used together for Cu leaching from PCBs. The overall reaction for copper dissolution in the mixture (HCl + H₂O₂) is given below [68],

$$Cu + H_2O_2 + 2HCl \rightarrow CuCl_2 + 2H_2O_2$$

Recently, a similar investigation was conducted by [33], which focused mainly on copper leaching and was followed by electro-winning process (Figure 1b). In contrast to the earlier study, mentioned in the above paragraph, here the authors used sulfuric acid (H₂SO₄) in the presence of H₂O₂ as an oxidant. The optimum concentrations were: H₂SO₄ (3.6 mol L⁻¹) and H₂O₂ (6% v/v), temperature (20 °C), pulp density (75 g L⁻¹), the leaching time was 186 min, and the leaching efficiency of Cu reached 96%. The electrowinning process allowed the recovery of over 92% of Cu from leach solution having a purity of 99.9%.



Figure 1. Cu metal recovery from waste PCBs. The flow sheet is drawn following information collected from (**a**) [31], (**b**) redrawn following [33].

Alkaline leaching agents have been used for metal leaching from PCBs. A recovery of 92% of Cu and 50% of Zn from PCBs was observed by using ammonia (NH₄OH), along with hydrogen peroxide (H₂O₂) [39]. These extraction values were obtained under the following conditions: 2 M NH₄OH, 17.5 M H₂O₂, at 40 °C and 400 rpm agitation.

In addition to strongly acidic and alkaline leaching agents, some researchers have used chemicals that can behave like acid or alkali to leach metals from PCBs. An amino acid such as glycine has been used for extracting Cu from PCBs under both acidic and alkaline leaching environments [36,40]. This reagent can act as an acidic or basic agent because it includes a carboxyl group (COOH) which is considered as an acidic group and also an amino group (NH_2) which is a basic group [69]. Based on these properties, a novel method was proposed for Cu leaching from PCBs with a green amino acid (glycine) in the presence of H_2O_2 [36]. The Cu leaching efficiency reached 94.08% in 8 h at 30 °C with 1 mol/L of glycine concentration, 10% H₂O₂, 1:100 solid–liquid ratio and 400 rpm of stirring speed. In yet another study, glycine was used in an alkaline environment as a sustainable approach for Cu recovery from PCBs [40]. The authors proposed a two-step alkaline leaching process by simply using glycine for base metal leaching, and a mixture of glycine and sodium cyanide (NaCN) for precious metal leaching in the first and second stages, respectively. The recoveries of base metals were 80.9%, 99.1%, 85.6%, 72.5% and 6.5% for Zn, Al, Pb, Cu and Ni, respectively. A similar study indicated that it is possible to recover about 96.5% Cu from PCBs by using alkaline glycine [50].

2.1.2. Metal Leaching from Liquid Crystal Display (LCD) Panels

Hydrometallurgical processes have been used as an important tool in several researches that were recently carried out for metal leaching from liquid crystal display (LCD) panels. The majority of these studies focus on the recovery of indium from LCD [27,57–59]. It is important to note that indium is basically and primarily recovered as a by-product of zinc through the processing of base metal concentrates [70]. The U.S. Customs and Border Protection Agency has included indium in the list of its commercial base metals [71]. Indium (In) is primarily used as indium tin oxide for the making of transparent conductive coatings for LCDs. The composition of LCD panels varies according to the variety, as well as the manufacturer and majorly consists of silicon, aluminium and calcium, which compose the glass of LCD panels. A range of other metals such as Zn, Cu and Fe can also be present in LCD. However, indium is considered as the most desired metal since it occurs in a higher amount in LCD waste. The studies conducted on the leaching of In from LCD panels in the last decade have been reviewed and can be found elsewhere [43,44,72].

Nevertheless, during the last three years, some studies have focused toward the improvement of In leaching from LCD. Several reports have indicated sulfuric acid as a suitable inorganic acid lixiviant due to its high selectivity and dissolution of indium oxide (In_2O_3) and tin oxide (SnO_2) from LCD [53,56–58] according to the following reactions [59].

$$\begin{split} In_2O_3 + 3H_2SO_4 &\to 2In^{3+} + 3SO_4^{2-} + 3H_2O\\ SnO_2 + 2H_2SO_4 &\to Sn^{4+} + 2SO_4^{2-} + 2H_2O \end{split}$$

In one of the studies, [57] used the Box–Behnken-type experimental design methodology to optimize and evaluate the effect of acid concentration (H_2SO_4), leaching time and temperature in In leaching from LCD panels. From their investigation, it was concluded that H_2SO_4 and temperature were the main parameters that influenced the metal solubilisation. Under optimum conditions (0.4 N H_2SO_4 ; 30 min; 70 °C and 50% of S/L ratio), about 99.5% of In was leached out. In a report made by [58], the leaching of In was carried out by using 0.5 M of H_2SO_4 at a solid/liquid ratio of 0.1 g/mL, 360 rpm and 90 °C for 2 h. Under these conditions, the leaching efficiency of In was about 98%.

Inorganic acid such as citrate can also be used for efficient In leaching in the presence of an oxidizing agent. However, since LCD panels contain Fe and Sn in addition to In, this reagent can lose its selectivity by co-dissolution of all of these metals [27]. In order to improve the efficiency and selectivity of In leaching, prior to citrate leaching, pre-treatment of LCD sample was done so as to remove the excess of Sn and Fe [27]. As a result, this allowed the recovery of approximately 99% In in 16.6 h by using 1M citrate and 0.2 M N₂H₄ as an oxidant at pH 5 and solid/liquid ratio of 20 g/L. However, compared to the earlier studies [57,58], it was observed that the leaching time was considerably higher and needed to be improved.

Of late, several researches were diverted towards the leaching of more than a single metal from LCD panels. As per a report made by [56] using 1 mol/L H₂SO₄ at 70 °C for 1 h, 99.25% of In could be recovered and 97.07% of Sn got precipitated in the solid form by the adoption of a single leaching step. The precipitation of Sn in the leaching process was due to the hydrolysis and precipitation of Sn⁴⁺ at pH < 1, according to the reaction shown below. The pregnant leach solution from the previous step was subjected to sequential electrodeposition for Cu and In extraction in the first and second steps, respectively.

$$\operatorname{Sn}^{4+}(\operatorname{aq}) + (2+x)H_2O \rightarrow \operatorname{SnO}_2 \cdot xH_2O_{(s)} + 4H^+(\operatorname{aq})$$

Leaching time can be reduced by using a mixture of H_2SO_4 and H_2O_2 as an oxidant. Recently, 3 M of H_2SO_4 with H_2O_2 was used to recover 86.3% of In and 76.1% Sn within a time period of 10 min at 85 °C [59]. H_2O_2 was used to enhance In and Sn dissolution during the leaching process. It was observed that In and Sn recovery increased with increasing concentration of H_2O_2 , while the HCl concentration was kept constant. The mechanisms involved in HCl leaching of In_2O_3 and SnO_2 from LCD are presented in the following chemical reactions below [55].

$$In_2O_3 + 6HCl \rightarrow 2InCl_3 + 3H_2O$$
$$InCl_3 + 6H_2O \rightleftharpoons [In(H_2O)_6]^{3+} + 3Cl^{-1}$$
$$SnO_2 + 4HCl \rightarrow SnCl_4 + 2H_2O$$

HCl is considered as a corrosive lixiviant [73]. This reagent can be replaced by weak acids for co-leaching of metals from LCD panels. A study carried out by [27] showed that citrate is capable of co-dissolving Fe, Sn and In by forming metal complexes. The results of the study indicated that Fe-citrate was thermodynamically more stable in the leaching

environment followed by Sn-citrate due to the high consumption of citrate by Fe and Sn, whereas the In-citrate complex was less stable.

2.1.3. Metal Leaching from Spent Batteries and Solar Cells

Several hydrometallurgical processes are used for leaching of different metals from spent battery based on the type of product and composition [44]. In these processes inorganic acids such as hydrochloric acid (HCl), sulfuric acid (H₂SO₄), nitric acid (HNO₃) and phosphoric acid (H₃PO₄) or organic acids like citric acid, oxalic acid, and tartaric acid were investigated for metal leaching from spent batteries [41]. Additionally, alkaline lixiviants such as ammonium hydroxide (NH₄OH), ammonium oxalate ((NH₄)₂C₂O₄), ammonium bicarbonate (NH₄HCO₃) and ammonium carbonate ((NH₄)₂CO₃) were also used for metal leaching from spent batteries [60].

Hydrometallurgical approaches have long been applied for metal extraction from alkaline batteries. In one of the studies, the extraction of base metal recovery from spent zinc-carbon and alkaline battery mixtures was investigated [62]. The work consisted of a stepwise procedure, where the first step included phosphorous removal by neutral leaching and it was subsequently followed by selective sulfuric acid leaching, purification and metal recovery processes. In this approach, 99.2% of Zn and 37.6% Mn were leached at 60 °C, pH 2 and liquid/solid ratio of 10. The whole flow sheet of the work is shown in Figure 2. A similar investigation was conducted for Zn and Mn leaching and recovery from spent alkaline batteries using sulfuric acid [74]. In yet another study, an alkaline glycine leaching was considered for Cd recovery from spent Ni-Cd battery. The findings of this study indicated that 86% of Cd could be selectively leached using 1.2 M of glycine at pH 9.6 and 25 °C for a time period of 150 min [63]. Moreover, HNO₃ was used to carry out Cd leaching from Ni-Cd battery. It was observed that 96.5% of Cd could be leached out in 180 min using 5 M HNO₃ at 70 °C [64].



Figure 2. Chemical leaching flow sheet of Zn and Mn extraction from alkaline battery (redrawn and adapted with modifications from [62].

With advances in studies and certain limitations for alkaline batteries, studies on Li-ion batteries (LIBs) have gained momentum. In case of LIBs, HCl, H₂SO₄ and HNO₃ are

$$2\text{LiCoO}_2 + 8\text{HCl} \rightarrow 2\text{CoCl}_2 + \text{Cl}_2 + 2\text{LiCl} + 4\text{H}_2\text{O}$$

 $2LiCoO_2 + 3H_2SO_4 + H_2O_2 \rightarrow 2CoSO_4 + O_2 + Li_2SO_4 + 4H_2O_2 + CoSO_4 + O_2 + CoSO_4 + 2H_2O_2 + 2H$

In a study carried out by [60], the effect of the concentration of H_3PO_4 , temperature, leaching time and liquid/solid ratio on the selective leaching of Li and Mn from a carbothermic reduced spent cathode of LIBs was studied. Using 2.75 mol/L H_3PO_4 at 40 °C, the leaching efficiencies of Li and Mn were 99.1% and 96.3%, respectively, within 10 min. The reactions involved in such process are illustrated below:

 $2H_3PO_4(aq) + MnO_{(s)} \rightleftharpoons Mn^{2+}(aq) + 2H_2PO_4^{-}(aq) + H_2O$

$$2H_3PO_4(aq) + Li_2CO_{3(s)} \rightleftharpoons 2Li + (aq) + 2H_2PO_4^{-}(aq) + CO_{2(g)} + H_2O_{3(g)}$$

A similar procedure was investigated by [38] for the leaching of Li, Mn, Co and Ni from spent electrical vehicle (EV) power batteries. The study involved the roasting of the material by sulfation, subsequently followed stepwise leaching experiments with water and H_2SO_4 , respectively. In the first stage, the roasted product was leached using water at 30 °C, 4 mL/g of L/S ratio for 2 h, which resulted in Li extraction of 90%. In the second stage, the residue from the first step was leached with 2 mol/L H_2SO_4 at 60 °C, L/S ratio of 5 mL/g for 2 h. During this last step 98% Ni, 97% Co and 90% Mn was leached from the residue.

Lately, solar cells are becoming a widespread and important source of green energy. They contain several valuable metals such as cadmium (Cd), chromium (Cr), lead (Pb), silver (Ag), selenium (Se) and tellurium (Te), copper (Cu), manganese (Mn), zinc (Zn) [75,76], titanium (Ti) and antimony (Sb) [77] based on the type of product. The production of waste solar cells is slow in comparison to other e-wastes due to their long lifetime (25–30 years) [76]; however, it is expected to increase in the near future [65]. Nevertheless, some hydrometallurgical studies are being conducted for their recycling aimed at metals extraction. Lixiviants, such as HNO₃ [65,78–81] and organic acid [82], are used for Ag leaching, while NaOH [64,76] and KOH [79] are investigated for Al leaching from solar cells.

In the study carried out by [79], a two-step chemical leaching process was applied after a pre-treatment process in order to leach out metals from photovoltaic modules. The first leaching step consisted of Ag dissolution using 3 M HNO₃, followed by leaching with 3 M NaOH for Al and Ti removal. These two processes allowed obtaining a final product of Si (99.98%). The same approach was applied recently by [65]. According to the study, 4 M HNO₃ was used at 80 °C for 4 h hours in the first step; whereas, in the second step, the same concentration of NaOH was used at 70 °C for 3 h. Leaching efficiencies for each step were 99.7% of Ag and 99.9% of Al, respectively. A typical metal leaching procedure for solar panels is illustrated in Figure 3.



Figure 3. A typical flow sheet for metal leaching from solar panels (Drawn following information from [65,79].

2.1.4. Metal Leaching from By-Product of E-Waste Sources

Electronic and electric equipment contain diverse elements that can be treated together or separately. During the pre-treatment or pyro-metallurgical processing of e-waste, some by-products such as flue dust [30] and sludge [51,52] may be generated. These by-products are mostly rich in valuable metals that need to be reprocessed. Flue dust from this source contains valuable metals such as Cu, Fe and Al in a ratio of 6%, 27% and 4%, respectively. Cu is generally found in the form of Cu metal, CuO and Cu₂O and Fe occurs as Fe₃O₄ [30].

As per a report made by [30], Cu and Fe could be leached out from the fine product (flue dust) produced by e-waste processing plant. During this investigation, two leaching steps (Figure 4) were proposed under fixed parameters of 20 g/L pulp density and 200 rpm stirring speed. The first step consisted of selective leaching of Fe with 1 M H₂SO₄ at 60 °C within 4 h; whereas, in the second step, 1 M HNO₃ was used for Cu leaching using the same temperature and leaching time. The metal recoveries were 90% of Fe and 98% of Cu for the first and second leaching stages, respectively. The proposed stepwise procedure seemed to be suitable, since under these conditions Fe was efficiently leached out as compared to Cu, which remained almost insoluble under low concentration of 1 M H₂SO₄. This could be due to the almost stable Gibbs free energy of metallic Fe (-78.364 kJ/mol) compared to metallic Cu (26.283 kJ/mol) according to the following reactions, respectively.

$$\label{eq:Fe} \begin{split} & Fe+H_2SO_4(aq) \rightarrow FeSO_4(aq)+H_{2(g)} \\ & Cu+2H_2SO_4(aq) \rightarrow CuSO_4(aq)+2H_2O_{(l)}+SO_{2(g)} \end{split}$$



Fine flue dust from E-Waste



Apart from flue dust, sludge generated by the PCBs manufacturing plant is also a source of metals, that is mainly composed of $Cu(OH)_2$ and $Fe(OH)_3$ [52]. The hydroxide form is generated due to the treatment of the wastewater by simple alkali precipitation [50]. Strong acids such as HCl, HNO₃ and H₂SO₄, have been tested in order to selectively leach out Cu from this secondary resource. Sulfuric acid was found to be the most suitable lixiviant in comparison to the rest [51,52,83]. The reaction involved in the dissolution of Cu(OH)₂ in the presence of sulfuric acid is given below [51,52].

$$Cu(OH)_2 + H_2SO_4 \rightarrow CuSO_4 + 2H_2O$$

As per reports [51], it is possible to leach out more than 96% Cu from the sludge of PCBs using 0.84 M sulfuric acid, L/S ratio of 100:1, 200 rpm, at a temperature of 60 °C within 80 min. In the study carried out by [52], Cu was selectively leached out from waste sludge generated from PCBs manufacturing process. Tests conducted with three different acids (HCl, HNO₃ and H₂SO₄) revealed that sulfuric acid showed the best Cu leaching efficiency. Under optimum conditions, (0.2 M H₂SO₄, 4% pulp density, 25 °C, 250 rpm, 60 min) the Cu leaching reached 95%. The study showed that a high acid concentration of 0.5 M led to the complete co-dissolution of Cu and Fe.

2.2. Bioleaching

The fast depletion of natural resources has motivated researchers and industrialists to exploit e-wastes for the recovery of metals in an eco-friendly and energy feasible manner. The biological method is similar to the chemical leaching route, except for it utilizes the reagents generated by microbes for metal extraction [84]. In addition, it is also considered to be an economically feasible and eco-friendly approach with higher efficacy, safety and easier management [85,86]. Several diverse microbial groups are involved in the leaching process including bacteria, fungi and yeast. These are specialized microbes and can be naturally isolated from mine sites or acid mine drainage samples [87].

The acidophiles including sulphur oxidizing bacteria and iron-oxidizing bacteria have found extensive application for bioleaching [88]. Apart from that, several organic and inorganic acids secreted by microorganisms were used for metal recovery from different low-grade sources. For example, microorganisms belonging to the genus *Bacillus* and *Pseudomonas* have the ability to extract metals from non-sulphidic sources by using organic carbon as a source of energy and carbon. The metabolic by-products produced by the microorganisms in the form oxalic acid, formic acid, citric acid and other bioreagents assist in the metal extraction from e-waste [89].

2.2.1. Bioleaching Mechanisms for Valuable Metals Recovery

The bioleaching of metals from e-waste mostly involves three mechanisms, namely: acidolysis, redoxolysis and complexolysis. Acidolysis involves the protonation of oxygen atoms that are present on the surface of metallic compound. Several biogenic organic (citric, succinic, formic, gluconic, oxalic formic and pyruvic acids) and inorganic acids (H_2SO_4) generated by heterotrophs are capable of carrying out acidolysis [90,91]. Redoxolysis involves oxidation-reduction reactions for metal solubiliation and the energy transfer essential for the growth of the microbe is derived through electron transfer. The complexolysis mechanism is mostly used by fungi and cyanogenic bacteria, in which cyanide is generated in the late stationary phase through the decarboxylation of glycine [85]. A mechanism showing the microbial mode of action for e-waste is shown in Figure 5.



Figure 5. Microbial mechanisms involved in leaching of metals from e-waste.

The bioleaching process for recovering metals from e-waste is mostly carried out with fungi and bacteria and it is dependent on the culture media composition, system pH and particle size of the waste material. Several microorganisms including *Aspergillus niger* (*A. niger*), *Aspergillus nominus* (*A. nominus*), *Acidithiobacillus thiooxidans* (*At. thiooxidans*) are reported for the recovery of base metals and precious metals such as Cu, Au, etc. [92]. Tables 2–4 lists such microbial studies on some selected e-waste streams. It is to be kindly noted that the tables present base and some other non-ferrous metals (wherever applicable) in order to reflect the overall study.

Bacterial Mechanisms

According to studies carried out by some investigators on the bioleaching mechanism of bacteria using *Acidithiobacillus ferrooxidans* (*At. ferrooxidans*) and *At. thiooxidans*, it is observed that no contact needs to be established between the bacteria and e-waste in order to ensure or initiate the bioleaching process [93]. In this process, the oxidation reaction (conversion of Fe^{2+} to Fe^{3+}) takes place by inorganic acid and enzymes. The reaction rate is enhanced by the production of H⁺ ion during the oxidation process [94]. Nevertheless, direct bioleaching of metals from e-waste utilizing *At. ferrooxidans* and *At. thiooxidans* is also reported [92,95]. The efficacy of the bioleaching process relies on the Fe²⁺ oxidation rate, concentration of Fe²⁺ ion and pH of the medium [92]. However, the direct mechanism of

bioleaching is yet to be understood completely. The maintenance of high redox potential in the leaching medium due to bacterial action is presumed to facilitate the indirect leaching. Since e-waste is devoid of any energy source, therefore, iron and sulfur are generally added to the medium in the form of ferrous sulphate and elemental sulphur.

The bioleaching process generally consists of the following steps: (a) oxido-reductive reactions, (b) the secretion of organic and inorganic acids and (c) the release or excretion of microbial metabolic products, complexing agents and chelators [89]. The redox reactions take place in the exopolysaccahride layer of the bacteria and the exopolymeric substances such as proteins, amino acid, lipids, etc., secreted by the bacteria play a vital role in the leaching process [89,96]. The microbe-substrate interactions hold prime importance in determining the efficiency of bioleaching. The reactions facilitating bioleaching of metals (combined acidolysis–redoxolysis) are shown in reactions below:

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

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

Chemical: Cu° (e.g., in e-waste) + 2Fe³⁺ (biogenic) $\rightarrow Cu^{2+} + 2Fe^{2+}$ (3)

$$Cu^{\circ}$$
 (e.g., in e-waste) + H₂SO₄ (biogenic) + 0.5O₂ \rightarrow Cu²⁺ + SO₄²⁻ (4)

Table 2. A list of bioleaching studies on some selected e-wastes using mesophilic microorganisms their metal recovery aspects. (Note: Metals like Li, In etc. have been included, wherever applicable, to reflect the overall study along with base metal dissolution).

	Microorganisms		Growir	ng Conditions			Optimum Bioleaching Conditions						
E-Waste		Cell Con.	pН	Temp.	Stirring Rate	Cell Con.	pН	Pulp Density	Temp.	Stirring Rate	Time	Recovery	References
	Acidithiobacillus ferrooxidans	10%	NA	30 ° C	170 rpm	5%	3	20 g/L	30 ° C	170 rpm	20 days	100% Cu and Ni	[97]
	Acidithiobacillus ferrooxidans	10%	NA	30 ° C	170 rpm	5%	1	8.5 g/L	30 ° C	170 rpm	17 days	100% of Cu and Ni	[98]
	Acidithiobacillus ferrooxidans	NA	1.75	35 ° C	150 rpm	10%	1.75	10 g/L	30 ° C	150 rpm	6 days	94% Cu	[99]
- - - PCBs -	Acidithiobacillus ferrivorans and Acidithiobacillus thiooxidans	5% (v/v)	2.5	30 ° C	150 rpm	$\begin{array}{c} 1.2\pm0.4\times10^8\\ CFU/mL \end{array}$	1.0–1.6	10 g/L	$23\pm2~^{\circ}C$	150 rpm	7 days	98.4% Cu	[100]
	Acidithiobacillus ferrooxidans	5% (v/v)	1.5	30 ° C	180 rpm	5%	1.5	18 g/L	30 ° C	180 rpm	64 h	94.1% Cu	[101]
	Acidithiobacillus ferrooxidans	10%	2	30 ° C	165 rpm	10%	2.25	2 g/L	30 ° C	160 rpm	78 h	92.57% Cu	[82]
	Acidithiobacillus ferrooxidans and Acidithiobacillus thiooxidans	NA	1.5– 2.0	30 ° C	150 rpm	NA	1.5	10 g/L	30 °C	150 rpm	7 days	95% Cu	[102]
	Acidiphilium acidophilum	NA	3.5	30 ° C	150 rpm	NA	2.5	1 g/L	30 °C	170 rpm	60 days	79% Cu, 29% Zn, 10% Pb, 39% Ni	[89]
	Acidithiobacillus ferrooxidans	$\frac{1\times 10^9}{cells/mL}$	2.5	30 ° C	170 rpm	NA	2.5	7.5 g/L	30 ° C	170 rpm	18 days	94% Cu, 92% Zn, 64% Pb, 81% Ni	[103]
	Bacteria consortium dominated by <i>Leptospirillum</i> <i>ferriphilum</i>	10%	1.7– 1.9	30 ° C	150 rpm	NA	1.8	10 g/L	$30\pm2~^{\circ}C$	150 rpm	2–4 days	>99% Cu, 29% Zn, 58% Ni	[104]
	Leptospirillum feriphillum	10%	2.0	$30\pm2\ ^{\circ}C$	150 rpm	10%	2	10 g/L	$30\pm2\ ^{\circ}C$	150 rpm	<4 days	>95% Cu, Zn, Ni	[105]
	Acidithiobacillus ferrooxidans	10%	NA	30 ° C	130 rpm	10%	2	15 g/L	30 °C	130 rpm	11–14 days	99% Cu (11d), 98% Ni (14d)	[106]
-	Acidithiobacillus ferrooxidans, Leptospirillum ferrooxidans and Acidithiobacillus thiooxidans	10%	1.8	30 °C	150 rpm	10%	1.8	10%	30 °C	150 rpm	8 days	98.1% Cu, 55.9% Al, 66.9% Zn, 79.5% Ni	[107]
	Acidithiobacillus ferrooxidans	10%	1.2	35 °C	250 rpm	NA	0.6-1.2	1%	25 °C	200 rpm	2 days	86.17% Cu, 100% Al, 100% Ni, 100% Zn	[49]
	Leptospirillum ferriphilum and Sulfobacillus benefaciens	10% v/v	1.3	35 °C	NA	NA	1.5	1% (w/v)	36 °C	600 rpm	2 days	96% Cu, 73% Ni, 85% Zn, 93% Co	[108]

			Growing Conditions					Optimum Bioleac	hing Conditior	IS		— Metal	
E-Waste	Microorganisms	Cell Con.	рН	Temp.	Stirring Rate	Cell Con.	рН	Pulp Density	Temp.	Stirring Rate	Time	Recovery	References
LCD _	Acidothiobacillus ferrooxidans and Acidothiobacillus thiooxidans	NA	NA	30 ° C	NA	10%	1.9	2.5% (w/v)	30 °C	NA	14 days	90.2% Sn	[109]
	Acidithiobacillus thiooxidans	10%	2	30 ° C	170 rpm	10%	2.6	1.6% (w/v)	30 °C	170 rpm	15 days	100% In, 10% Sr	[110]
Zn-Mn Batter- ies	Acidithiobacillus ferrooxidans	5%	2	30 °C	140 rpm	NA	2	10 g/L	30 ° C	140 rpm	21 days	99% Zn, 53% Mn	[111]
	Acidothiobacillus thiooxidans	10% v/v	4.5	30 ° C	200 rpm	NA	2.4	0.25%	30 °C	200 rpm	40 days	22.6% Co, 66% Li	[112]
LIBs	Acidithiobacillus ferrooxidans,	10%	2	30 ° C	160 rpm	10%	1.93	100 g/L	30 °C	160 rpm	3 days	90% Ni, 92% Mn, 82% Co, 89% Li	[113]

Table 2. Cont.

Table 3. A list of bioleaching studies using moderately thermophilic microorganisms and their metal recovery aspects fromselected e-wastes. (Note: Li is included, wherever applicable, to reflect the overall study along with base metal dissolution).

			Growing Conditions					Optimum Biolead	ching Condition	ns		Metal	
E-Waste	Microorganisms	Inoc.	рН	Temp.	Stirring Rate	Cell Con.	рН	Pulp Density	Temp.	Stirring Rate	Time	Recovery	References
E-Scrap	Sulfobacillus thermosulfdooxidans and Thermoplasma acidophilum	10%	NA	45 °C	180 rpm	10% w/v	2	10% w/v	45 °C	180 rpm	12 days	90% Cu, 80% Al, 82% Ni, 85% Zn	[114]
PCBs	Sulfobacillus thermosulfdooxidans	NA	1.75	50 °C	150 rpm	10%	1.75	10 g/L	50 °C	150 rpm	6 days	99% Cu	[99]
	Leptospirillum ferriphilum and Sulfobacillusthermo- sulfdooxidans	10%	0.9	42 °C	200 rpm	10%	0.9	100 g/L	32 °C	180 rpm	9 days	93.4% Cu	[115]
LIBs	Leptospirillum ferriphilum sp. and Sulfobacillus thermosulfidooxidans spp.	10%	1.2	42 °C	180 rpm	10%	1.2	15 g/L	42 °C	180 rpm	3 days	100.0% Li, 99.3% Co,	[116]
	Leptospirillum ferriphilum and Sulfobacillus thermosulfidooxidans	10%	1.25	42 °C	180 r/min	NA	1.25	5 g/L	42 °C	180 r/min	1.5 days	98.1% Li, 96.3% Co	[117]

Table 4. A list of bioleaching studies on some selected e-wastes using heterotrophic microorganisms and their metal recovery aspects. (Note: Au, Ag, Li, In, etc., are included, wherever applicable, to reflect the overall study along with base metal dissolution).

E Masta	Microorganism	Crowth Modia	Energy Source		Optim	um Bioleaching Cond	litions		 Metal Recovery 	Poforoncoc	
E-waste	witcioorganism	Growth Media	Energy Source =	рН	Pulp Density	Temp	Stirring Rate	Time	- Metal Recovery	Kererences	
	Chromobacterium violaceum	LB medium	0.5 g glycine	7.2	1% w/v	30 °C	150 rpm	7 days	79% Cu, 46% Zn, 9% Fe, 69% Au, 7% Ag		
	Chromobacterium violaceum and Pseudomonas aeruginosa	LB medium	0.5 g glycine	7.2.	1% w/v	30 °C	150 rpm	7 days	83% Cu, 49% Zn, 13% Fe, 73% Au, 8% Ag	[118]	
	Bacillus subtilis and Bacillus cereus	NA	NA	6–8	10 g/ 150 mL	37 °C	120 rpm	25 days	48% Zn, 93% Cd	[119]	
PCBe	Bacillus megaterium	Nutrient broth medium	0.5 g/L glycine	10	2 g/L	30 °C	170 rpm	10 days	13.26% Cu, 36.81% Au	[120]	
1003	Aspergillus niger	PDA	50 g/L glucose	4.4	NA	28 °C	280 rpm	14 days	29% Cu, 87% Au	[121]	
	Aspergillus niger	PDA	100 g L ⁻¹ sucrose	NA	0.5–20.00 g L ⁻¹	Ambient temp	120 rpm	21 days	100% Zn, 80.39% Ni, 85.88% Cu	[122]	
	Aspergillus niger	PDA	20 g Dextrose	5.0	$2\mathrm{gL}^{-1}$	17–24 °C	NA	35 days	2.8% Cu, 0.53% Au	[123]	
	Streptomyces albidofavus	ISP 2 broth medium	NA	NA	1.5%	28 °C	120 rpm	5 days	66% Al, 74% Ca, 68% Cu, 65% Cd, 42% Fe, 81% Ni, 82% Zn, 46% Pb	[124]	
LIBs	Aspergillus niger MM1	Sucrose medium	100 g/L sucrose	6.	0.25%	30 °C	200 rpm	40 days	82% Co, 100% Li	[112]	
Ni-Cd Batteries	Aspergillus niger	RB medium	NA	NA	0.3 g/80 mL	28 °C	150 rpm	21 days	81.41% Ni, 92.31% Cd	[125]	

E-Waste	Missoanniam	Growth Media	Energy Source —		Optimu	Matal Recovery	D. (
	witcroorganism			рН	Pulp Density	Temp	Stirring Rate	Time		Kererences
LCD	Aspergillus niger	PDA	100 g/L Sucrose	4.0	1% (w/v)	70 °C	125 rpm	90 min	100% In	[126]
AMOLED Displays	Bacillus foraminis	TSA and TSB	15% glycerol	7.7	7%	40 °C	160 rpm	12 days	56.8% Mo, 41.4% Cu, 100% Ag	[127]
Solar Cells	Penicillium chrysogenum	Sucrose medium	100 g/L sucrose	NA	1% (w/v)	20 °C	200 rpm	3 days	100% Te, 98% Al	[128]

Table 4. Cont.

Fungal Mechanism

The precious metal recovery from e-waste is generally instigated by biogenic cyanide or organic acids produced by fungi. The organic acids (natural chelating agents) generally act as complexing agents and mostly include oxalic acid, tartaric acid and citric acid. Reports indicate the use of *A. niger* for the recovery of Au and Ag from e-waste, where organic acids such as gluconic acid and citric acid are produced by the microbe for carrying out the leaching process. The reaction as a result of the microbial (*A. niger*) action is shown below:

$$C_6H_{12}O_6 + 1.5O_2 \rightarrow C_6H_8O_7 + 2H_2C_7$$

The leaching operations can take place under both aerobic and anaerobic conditions and may be influenced by temperature, solid-liquid ratio and pH of the system [92].

2.2.2. Bioleaching of Metals from Waste Printed Circuit Boards (PCBs)

Bacterial assisted leaching of scrap TV circuit boards was carried out for the recovery of copper using a mixed culture of mesophilic bacteria (comprising of At. ferrooxidans, L. ferrooxidans, At. thiooxidans) [129]. The bioleaching process was seen to be dependent on ferrous iron oxidation efficiency of the bacteria and the initial availability of soluble iron in the medium. Lab-scale shake flask experiments were conducted using At. ferrooxidans for copper extraction from printed circuit boards [130]. Recently, mechanical activation was proposed by [131] in order to enhance the bioleaching efficacy of At. ferrooxidans for metal extraction from waste printed circuit boards (WPCBs). The leaching rates of Cu, Ni and Zn could be enhanced by using mechanical activation, which was approximately 20% higher in comparison to the un-activated WPCBs. Additionally, in one of the studies, a direct DC electric field was applied for enhancing bioleaching and copper recovery from e-waste in a bioelectric reactor. The DC electric field enhanced the bacterial growth and activity, simultaneously facilitating the ferrous iron oxidation and resulting efficient leaching of copper from printed circuit boards (PCBs) [132]. Apart from At. ferrooxidans, a pure culture of the acidophilic bacteria, Acidiphilium acidophilum (A. acidophilum), was also used for the bioleaching of specific metals such as Cu, Zn, Pb and Ni from e-waste [103]. In addition, heterotrophic species such as Acinetobacter sp. CrB₂ was used for the bioleaching of copper from e-waste. The copper bioleaching occurred as a result of the combined action of extracellular enzymes and metabolites produced by the bacteria and increasing the number of cycles of operations increased the bioleaching efficiency [133]. Of late, some indigenous cyanogenic bacterial strains isolated from e-waste landfills have been also found to be potential microorganisms for copper extraction from e-waste under optimized conditions [134]. Apart from that, lately a Bacillus sp. isolated from Hymeniacidon heliophila sponge cells has shown the potential to leach copper from e-waste, where copper was produced in the form of copper nanoparticles. The peptides released by bacteria were responsible for leaching of copper, absorption of copper ion and their incorporation into cells for nanoparticle formation [135].

Besides bacterial strains, several fungal strains have been reported for metal extraction from e-waste. Fungi are capable of secreting large number of organic acids, amino acids and other metabolites which assist in metal dissolution. The fungal metabolites act by displacing metal ions with hydrogen ions or by developing soluble metal complexes and chelates, subsequently leading to metal dissolution [136]. Recently, *Penicillium simplicissimum (P.*

simplicissimum) was investigated for copper and nickel extraction from PCBs of mobile phones. Four different carbon sources such as sucrose, cheese whey, sugar, and sugar cane molasses were used for the study and it was observed that non-conventional carbon sources improved the bioleaching efficacy [137]. Apart from that, mixed fungal cultures were used for metal extraction from e-waste [138]. The gradual adaptation of microorganisms to heavy metals helps in the development of heavy metals tolerant microbes, which can be utilized for industrial scale applications [138,139]. Examples of such fungal strains include *Asergillus foetius* (*A. foetius*) and *Penicillium funiculosum* (*P. funiculosum*) [140,141]. More experimental works on WPCB bioleaching can be found in Tables 2–4, respectively.

2.2.3. Bioleaching of Metals from Spent Batteries

A variety of batteries such as lithium-ion batteries (LIBs) and alkaline batteries including nickel-cadmium, nickel-hydrogen [142], Zn-Mn batteries [111], and zinc-carbon batteries [62] exist in the market based on the models.

In case of alkaline batteries, it has been be found that autotrophic acidophiles have been applied for the leaching of base metals. For example, *At. ferrooxidans* has been used for metal recovery from Ni-Cd batteries [143–145]. *Alicyclobacillus* sp. and *Sulfobacillus* sp. [146], *At. thiooxidans* [147–149], *L. ferriphilum* [148] and *At. ferrooxidans* [111,113] have been used for metal recovery from spent alkaline Zn-Mn batteries. In a study conducted by [111], the culture supernatant of *At. ferrooxidans* was used for Mn and Zn dissolution from waste alkaline button-cell batteries. At an initial pH of 2, temperature of 30 °C and 10 g/L pulp density, 99% of Mn and 53% of Zn were extracted in 21 days. A similar investigation was conducted by [113] with the same strains of bacteria for metal dissolution from spent Ni, Mn, Co (NMC) batteries. The findings of this study showed that an improved leaching time (3 days) and pulp density (100 g/L), 90%, 92%, 82%, and 89% dissolution for f Ni, Mn, Co and Li, respectively, could be achieved.

Moreover, heterotrophic fungus has been also applied for metals extraction from this type of batteries. Kim et al. [139] showed that most of the *Aspergillus* species are able to dissolve metal from spent Zn-Mn and Ni-Cd batteries. This assertion was confirmed by [125] through a comparative study on bioleaching of nickel and cadmium recovery from spent Ni-Cd batteries by using *A. niger*. In this work, different culture media such as potato dextrose broth (PDB), malt extract broth (MEB) and Richards's broth (RB) were investigated. It appeared that the application of *A. niger* in RB medium in a two-step bioleaching process, resulted in efficient dissolution of both base metals (Ni—81.41% and Cd—92.31% in 21 days).

Apart from the alkaline batteries, the LIBs are now widely prevalent and studied. Several bioleaching studies have been undertaken on metal extraction from spent lithium-ion batteries (LIBs) by using autotrophic acidophiles [112,150] and heterotrophic microorganisms [112,139,151,152]. A comparative bioleaching study was investigated by using spent medium of *A. niger* (MM1 and SG1) (heterotroph) and *At. thiooxidans* 80,191 (autotrophic acidophile) for Co and Li leaching from spent LIBs [112]. It was observed that fungal bioleaching (spent medium of *A. niger* MM1) was more effective for metal dissolution from LIBs compared to bacterial leaching. The findings of the research indicated Co and Li dissolution of 82%100%, respectively.

The process is illustrated in Figure 6. Moreover, in order to increase the efficiency of bioleaching, the optimization of bioleaching parameters for metal extraction from LIBs was conducted by [152] using *A. niger*. Additionally, heterotrophic bacterial such as *Gluconobacter oxydans* (*G. oxydans*) [153] and another strains of *Aspergillus* like *A. nomius* JAMK1 [151] were proposed in the optimization of bioleaching parameters for metal recovery from LIBs.





Figure 6. Bioleaching of LIBs with A. niger (drawn based on information collected from [112]).

On the other hand, some studies have suggested efficient metal extraction from spent LIBs at higher pulp densities and shorter leaching time by using autotrophic meso-acidophilic bacteria such as *At. ferrooxidans*. A study carried out by [150] indicated that using this bacterium for bioleaching of LIBs at 40 g/L of solid/liquid ratio resulted in 100% of Li, 88% of Co, and 20% of Mn dissolution within 12 days. A consortium of meso-acidophile and moderate thermo-acidophile (*L. ferriphilum* and *S. thermosulfidooxidans*) was used to study the effects of higher pulp density on the leaching of waste LIBs. At 5 g/L of pulp density and 42 °C, 98.1% Li and 96.3% Co were leached in 1.5 days [117].

2.2.4. Bioleaching of Metals from LCD/LED Panels

Most of the studies carried out on the bioleaching of liquid crystal displays (LCDs) are focused on the recovery of indium. A brief description on indium application in LCD is presented in Section 2.1.2. Adapted acidophilic strains such as *At. ferrooxidans* and *At. thioxidans* were used for the extraction of indium and tin from LCD panels, where a maximum leaching of 55.6% and >90% was obtained for indium and tin respectively [109]. Apart from acidophilic strains, heterotrophic strains such as *A. niger* are used for the extraction of In from waste LCD panels through the optimization of the fermentation method. Through this method, In bioleaching efficiency increased from 12.3% to 100%. The carboxyl groups from organic acids and proteins were identified as the crucial factors for the release of H⁺ ions required for leaching of indium [126].

Aside from LCDs, LEDs (light emitting diodes) are receiving considerable interest as a secondary source for valuable metals. In addition, they pose a threat to the environment as a source of pollutant. Therefore, of late, studies have been carried out in this regard for the recovery of metals from LEDs. In one of the approaches, a novel stepwise indirect bioleaching technique was studied using adapted cells of *At. ferrooxidans*. The rate of bioleaching was improved by the stepwise addition of biogenic ferric and was maintained at 4-5 g/L. The results of the study revealed that the direct bioleaching approach, which involves bacterial attachment to the sample resulted in lower yields of metals. In contrast, the indirect bioleaching approach using biogenic ferric resulted in higher metal recovery from LEDs, where at a pulp density of 20 g/L, nearly 83% copper, 97% nickel and 84% gallium could be recovered. Metals such as nickel and copper have a bacteriostatic effect

on the acidophiles. This is seen to interfere with the enzyme catalysed oxidation of Fe and reproduction. Consequently, the lower yields with direct bioleaching are attributed to this effect [154]. It is also observed that adapted strains of *At. ferrooxidans* provide better resistance to the high concentration of metals present in LEDs. In a study carried out by [155], it was observed that the heavy metals tolerance of *At. ferrooxidans* reduced with increasing pulp density from 5 to 20 g/L. Nevertheless, the adapted cells had higher Fe³⁺ level, cell number, ORP and lower pH than the non-adapted cells, which resulted in better a leaching efficiency of metals, where 84%, 96%, and 60%, copper, nickel and gallium, respectively, could be extracted.

2.2.5. Bioleaching of Spent Solar Panels and Some By-Products of E-Waste Resources

The application of bioleaching on metal recovery from solar panels is very limited. Nevertheless, recently [128] have investigated the potential metal leaching abilities of four different microorganisms from solar panels which contain B, Mg, V, Ni, Zn, Sr, Cr, and Te. In the study, spent media of two autotrophic (acidophilic) bacteria (*At. thiooxidans, At. ferrooxidans*) and two heterotrophic fungi (*P. chrysogenum* and *P. simplicissimum*) wereused. It was observed that the spent medium of *At. ferrooxidans* was effective for Mg, Mn, Co, and Zn leaching. *At. thiooxidans* was capable of B and Zn dissolution, and *P. simplicissimum* was able to leach only Mg, whereas the spent medium of *P. chrysogenum* was able to dissolve almost all the aforementioned metals. Based on this comparative investigation, the authors concluded that *P. chrysogenum* spent medium was the most effective for multiple metal leaching from waste solar cells. Under conditions of 30 °C, 150 rpm and 1% (w/v) pulp density, the leaching efficiencies of B, Mg, V, Ni, Zn, and Sr was 100% and Te was 93%.

Bioleaching has been also applied for metal recovery from the by-product of e-waste. Yan et al. [156] investigated bioleaching of Cu and Ni from electroplating sludge by using *At. ferrooxidans* and *At. thiooxidans*. Additionally, [157] have developed two successive bioleaching steps for base metals, precious metals and REE extraction from the dusts generated by e-waste shredding process. In the first step, *At. thiooxidans* was used for base metals leaching (almost 100%) at pH 3.5 during 8 days, whereas, cyanide producing *P. putida* WSC361 was applied for gold dissolution (48%) within 3 h.

3. Integrated/Hybrid Approaches

Recent years have been witnessing the utilization of some hybrid approaches for enhancing the metals recovery from e-wastes. The rapid diminution in resources and the significant ecological footprints have compelled investigators to comb for green approaches for metal recovery. The hybrid methods provide a stepwise combination of different approaches in order to deliver an enhanced and resourceful system for metal recycling [158]. In a report made by [159], several ligand-microbe combinations (an example is shown in Figure 7) were proposed and described as hybrid techniques for the efficient extraction of the desired metals. In one of the investigations carried out by [103], the bioleaching of some specific metals such as Cu, Zn, Pb and Ni from high grade PCBs was studied using pure culture of At. ferrooxidans. The study was carried out in the presence and absence of lemon juice, which contains 0.2 M citric acid as an active constituent and natural tetradentate as a chelating agent, that provide a hybrid environment for enhanced metal recovery. Results of the study indicated enhanced metal solubilization under hybrid conditions, as a result of the chelating effect of citric acid and a maximum leaching of 94% Cu, 92% Zn, 64% Pb and 81% Ni was observed after 18 days, using a size range of 0.075–1 mm and pulp density of 7.5 g/L.



Figure 7. Pictorial representation of a ligand mediated integrated bioleaching approach.

Yet another study carried out by [158] demonstrated the use of a novel two-step biorecovery approach, subsequently followed by electrochemical treatment for the recovery of copper from waste PCBs. In the study, an isolated strain, USCT-R010 was used for copper leaching under optimized conditions and the leach liquor was then subjected to a purification step using biosorption technique, where dead biomass of *Aspergillus oryzae* and Baker's yeast were used as biosorbents under optimized conditions. Following desorption and electrowinning, 92.7% copper was recovered from the eluate and the characterization studies revealed that the recovered copper had 95.2% purity. This study indicated the utilization of cost-effective biomaterial towards metal purification and recycling while, at the same time, providing an efficient and ecofriendly approach for metal recovery. In addition, the hybrid technique is often less time consuming than the individual approach involving only bioleaching and allows the extraction of valuable metals from low grade ores, secondary wastes, etc.

Likewise, several researchers have reported that ferric sulphate is applicable for metal leaching from PCBs [49,160,161]. A study carried out by [161] revealed that 100 mM Fe₂(SO₄)₃ is able to leach more than 98% of Cu from PCBs at 20 °C, 300 rpm and 10 g·L⁻¹ of pulp density within 4 h. A similar study was conducted by [49]. In their investigation, the authors made a comparative study between chemical and biogenic ferric leaching. The purpose was to understand the function of these two sources of Fe(III) towards the leaching of Cu, Ni, Al and Zn from PCBs. Their findings confirmed that there was no major difference in the leaching efficiency of these metals with the use of either biogenic or chemical ferric sulfate. Under optimum conditions, the chemical leaching attained 84.3% of Cu extraction, 98.4% of Ni extraction and 100% extraction for both Zn and Al.

4. Challenges, Future Prospects and Conclusions

During the last few decades, various studies have been carried out on the management and recycling of electronic and electrical wastes. As discussed in the introduction section, this is due to the fact that mineral resources are becoming increasingly scarce and also due to the environmental problems associated with these wastes. Circular economy, involving the principle of the three Rs (reduce, reuse and recycle), is currently the focus for waste recycling and holds prime significance in the current scenario [17,162]. In this aspect, recently (April 2021), a voluntary certification scheme for waste electrical and electronic equipment (WEEE) and batteries treatment was proposed through the EU Horizon 2020 CEWASTE project [163].

Theoretically, the concept of the circular economy remains the ideal solution for the integrated management (collection and recycling) of electronic waste. However, its applications in certain aspects of extractive metallurgy present an enormous challenge with respect to the implementation of an economically profitable and eco-friendly technology [164–167]. The foremost challenges that are to be taken into consideration are the long-term availability of electronic waste as raw material for a metallurgical plant, their diversity (PCBs, batteries, LCDs, solar panels, etc.) and their metal content, including the metals of interest. From the technical aspect, the diversity of electronic wastes and their content play an important role in the choice of technology, i.e., hydrometallurgy and bio-hydrometallurgy in the current case. As it can be understood from all the above sections, the contributions of chemical and biological routes are quite effective; however, each process has its own merits and demerits. A list of such aspects is given in Table 5.

Mathad	Chamical Mathad	Piplopical Mathad				
Method	Chemical Wiethod	Biological Method				
Merits	 Short leaching time; High recovery rate; High selectivity compared with bioleaching; High pulp density. 	 Suitable for e-waste treatment from an environmental point of view (eco-friendly process); Low capital and operation costs; Less energy consumption; Simple process and easy to maintenance. 				
Demerits	 Used some toxic chemicals that can cause environmental problems; Causes corrosion problems due to the use of certain corrosive chemicals; High capital and operation costs compare with bioleaching; Produce hazardous by-products. 	 Long leaching time and low pulp density; Leach efficiency is most of the time low compared with chemical leaching; Need for a better understanding of the type of e-waste for a best choice of microorganism (heterotrophic or autotrophic); Microorganism can be contaminated due to the high concentration of heavy metals and other contaminants in some e-waste; Limited use for industrial processing of e-waste. 				

Table 5. The Merits and Demerits of Chemical and biological methods.

For both of these processes, it can be seen that many studies were undertaken for PCBs compared to the rest of e-waste streams. For example, there is limited information available on the metal extraction from solar panels and by-product of e-waste. Consequently, such waste streams need to be deeply investigated within the scope of future works. With respect to chemical leaching (hydrometallurgy), the use of green lixiviants should be practiced more or low-cost, environmentally friendly methods can be developed that can facilitate/promote multi-metal leaching. Though bioleaching (bio-hydrometallurgy) provides

more eco-friendly benefits and is considered economic in comparison to chemical methods, still the search for more robust microbes that can enable efficient metal dissolution from specific or wide varieties of e-wastes should be prioritized. In addition, bioinformatic platforms can also contribute to receiving more useful information related to key microbial species involved in bioleaching [168–170]. According to a study, a bioleaching bacterial protein finder system was proposed that can predict putative proteins and make an assumption regarding any microbe that is capable of iron and sulphur oxidation (a key aspect in bioleaching operations) [171]. More of such bioinformatic attempts should be made to identify potential bioleaching microbes and then integrate them with the applied aspects of bioleaching. This would allow further validation of the bioinformatic analysis through wetlab experimental findings. Modifications or upgrading the process engineering aspects can also be an appropriate way to improve performance in both the systems/routes. Moreover, integrated approaches (chemical and bioleaching) can be implemented and tested to monitor the overall process efficiency for treatment of any specific e-waste and subsequently studied on a pilot scale, with the aim of finding industrial applications. Nevertheless, such attempts should be validated through techno-economic feasibility analysis [172] and life cycle assessment (LCA) [173] of the processes, considering the environmental and social impacts.

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