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Journal of Material Cycles and Waste Management (2024) 26:1349–1368
https://doi.org/10.1007/s10163-024-01944-4

REVIEW

Recent chemical methods for metals recovery from printed circuit boards: A review

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Received: 26 October 2023 / Accepted: 24 March 2024 / Published online: 3 April 2024
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Abstract

As the volume of e-waste continues to rise, it is crucial to sustainably manage printed circuit boards (PCBs) and their valuable metal components. PCBs are ubiquitous in modern society, powering a variety of electronic devices. The metal resource crisis and the imperative for a low-carbon circular economy have accelerated the development of e-waste recycling technology. High-value discarded PCBs represent a vital component of e-waste. However, discarded PCBs are deemed hazardous to the ecosystem due to the presence of heavy metals and brominated organic polymers. Thus, recycling metals from discarded PCBs is not only a strategic necessity for fostering a green ecological civilisation but also a crucial guarantee for ensuring a safe supply of mineral resources. This comprehensive review gives the profound details of PCBs, and the performance of and advances in the latest chemical metal recovery methods. Reviewing the latest metal recovery processes, we explored the application of diverse leaching agents, including ionic liquids (ILs), deep eutectic solvents (DESs), organic acids and amino acids. These solvents were assessed in terms of their recovery efficiencies, and most of them demonstrated excellent leaching performance. The role of optimising leaching parameters such as concentration, oxidants, pH, particle size, solid-to-liquid ratios (S/L), temperature, and contact time is underscored, offering insights into achieving sustainable PCB recycling practices. Most of these recent leaching methods successfully extracted base metals (Cu, Fe, Zn, Sn, etc.), as well as precious metals (Au and Ag), achieving leaching efficiencies exceeding 90.0%. Interestingly, their effectiveness can compete with that of traditional hydrometallurgical methods.

Keywords Electronic waste · Ionic liquids · Deep eutectic solvents · Organic acids · Amino acids

Introduction

In recent times, electrical and electronic companies across the globe have profoundly influenced the world, making their products an indispensable part of everyday life [1]. Electronic appliances encompass various household essentials, including refrigerators, televisions, washing machines, smartphones, laptops, photocopiers, iPads and so on [2, 3]. As the use of these items continues to grow among the populace, the inevitable consequence is the generation of associated waste. Electronic waste (e-waste) refers to discarded or non-functional electronic devices that have outlived their utility [4, 5]. More precisely, e-waste can be defined as the entirety or components of electrical or electronic equipment that users discard as waste, as well as the remnants from the manufacturing, repair, and refurbishment stages of the production process [3, 6, 7]. According to one of the previous reports, about 42.0% of e-waste originates from household appliances, 34.0% from communication devices, 14.0% from electronic gadgets, and the remaining 10.0% from accessories [8].

Moreover, about four years ago, the global production of e-waste reached a staggering 53.6 million metric tons (Mt) [9]. In addition, projections by the E-waste Statistics Partnership indicate that by 2030, the annual global accumulation of discarded electrical and electronic equipment (DEEE) will surge to an estimated 74.7 Mt [9]. Unfortunately, the issue of excessive DEEE generation has swiftly

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evolved into an urgent concern on a global scale [10]. DEEE comprises a multitude of components that can pose detrimental effects on the environment as well as human health. Indiscriminate handling and informal recycling practices can escalate DEEE into a significant global concern for public health and the environment [11, 12]. On the other hand, owing to its physical attributes, DEEE is exceptionally well-suited for recycling, promising substantial benefits when subjected to sound environmentally benign recycling processes [13]. This dual nature of DEEE renders it a compelling secondary resource while simultaneously presenting environmental challenges [14]. Furthermore, discarded printed circuit boards (DPCBs) represent a vital component within the scope of DEEE. They are characterised by their substantial inclusion of both high-value resources (metals) and hazardous substances, making them a crucial target for DEEE disposal and resource recycling [15–17]. In terms of their overall composition, DPCBs typically encompass a diverse array of bare boards intermixed with numerous electronic components (ECs). These ECs found on DPCBs often include capacitors, relays, resistors, and integrated circuits, among others. It is important to note that the composition and concentration of these two categories of materials exhibit significant variability, further intensifying the intricacy of the recycling process [18]. A typical recycling process for DPCBs is depicted in Fig. 1.

Conventional methods for recovery of metals from DPCBs encompass mechanical, hydrometallurgical, and pyrometallurgical processes, as extensively explored in previous research [19–21]. However, these established methods are deemed unsustainable due to their high costs, the significant pollution they cause, and their substantial energy consumption, amidst other issues [22–24]. Biometallurgical processes harness microorganisms to extract metals from solid PCBs, converting them into soluble forms in an aqueous solution by generating lixiviants, which play a pivotal role in the metal extraction process [5, 25–27]. The use of such microorganisms holds significant promise because it is environmentally friendly, can operate at ambient temperatures and pressures, is cost-effective, and results in minimal secondary waste generation [28, 29]. However, the commercialisation of this approach faces challenges, primarily due to an insufficient scientific database, including optimal conditions, for scaling up the process and synchronising acidic and alkaline biometallurgical processes [5]. So, the biometallurgical process might not be able to replace traditional methods. Previous review articles have dealt with hydrometallurgy involving the use of traditional solvents such as H_2SO_4, HCl, HNO_3, aqua regia, and alkaline reagents, among others, for the recovery of metals from DPCBs [30–32]. Other authors have also presented their review publications on pyrometallurgy/pyrolysis and biometallurgical methods [15, 21, 33, 34]. Notably absent from these discussions, however, is the utilisation of recent green solvents such as ionic liquids (ILs), deep eutectic solvents (DESs), organic acids, and amino acids in metal leaching from DPCBs. Therefore, this review aims to fill this gap, offering a comprehensive and up-to-date assessment of metal recovery from DPCBs using these innovative solvents. By focusing on ILs, DESs, organic acids, and amino acids, it strives to analyse the current state of research, evaluate efficiency, and propose future research directions. In other words, this article discusses specific chemical approaches for metal recovery, including the utilisation of ILs, DESs, organic acids, and amino acids. The factors influencing the recovery of metals from DPCBs are explored in a dedicated section, addressing key considerations in the process. The article concludes by summarising key findings and providing

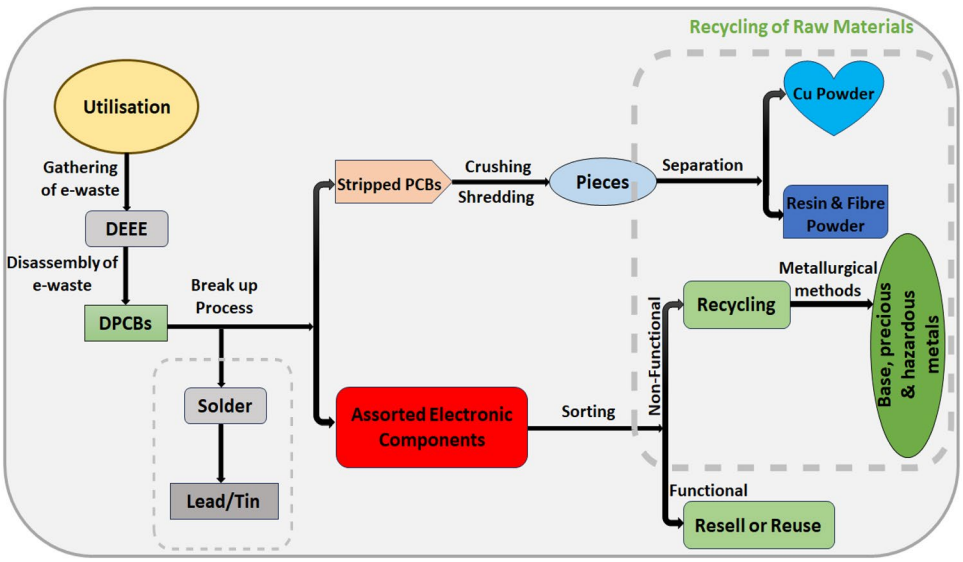


Fig. 1 Typical recycling flow chart for DPCBs in DEEE

insights into potential future research directions in the field of metal recovery from DPCBs. Synthesising knowledge from past studies, this review seeks to enhance our understanding of the potential of these alternative solvents in DPCB recycling. In doing so, it aspires to contribute to sustainable practices in e-waste management and the circular economy of valuable metals.

Recovery methods for metals from DPCBs

Traditional approaches for recovering metals from DPBCs have historically relied upon energy-intensive pyrometallurgical processes or the use of highly oxidative and acidic conditions in hydro- and solvometallurgical methods. These methods involve the utilisation of potent acids such as HCl, H_2SO_4, and HNO_3, as well as hazardous substances like aqua regia and piranha solution as earlier mentioned [35, 36]. Also, these methods may employ toxic oxidants like Cl_2, or even resort to cyanide (CN^-) and Hg, potentially resulting in the generation of substantial quantities of toxic and corrosive waste. The use of such "harsh" conditions can also contribute to reduced selectivity, thereby exacerbating the environmental impact [5, 37]. This approach contradicts the goals of sustainable development, which might appear paradoxical when considering recycling—an essential principle of the circular economy and green chemistry. The quest for safer, cost-effective, and environmentally friendly solvents, oxidants, reductants, ligands, additives, solid traps, and carriers is paramount to enhancing the sustainability of various metal recovery processes from DPCBs and other e-waste components. Therefore, in this section, we illuminated the use of ILs, DESs, organic acids and amino acids in the recovery of various metals from DPCBs. The summary of some of the research findings reviewed in this article is presented in Table 1.

Recovery of metals mediated by ionic liquids

ILs are a class of purely ionic, salt-like materials that are liquid at unusually low temperatures. The definition of ILs uses the boiling point of water as a point of reference: "ILs are ionic compounds which are liquid below 100.0 °C" [38, 39]. ILs have many potential applications owing to their ability to act as powerful solvents and electrolytes. Interestingly, ILs offer some great advantages over traditional leaching agents, such as acids or cyanides, for metal extraction from DPCBs and other components of e-waste or ores. ILs have high selectivity, low toxicity, low volatility, high stability, and an outstanding metal recovery ability [40–42]. Additionally, they can also be reused and recycled, thereby reducing waste generation and environmental impact. There are different methods of using ILs for metal extraction from e-waste. Such methods include leaching, liquid–liquid extraction, electrodeposition, and aqueous biphasic systems [41, 42]. However, our focus here is the leaching (i.e. solid–liquid extraction) of metals from DPCBs using ILs.

Based on the available information in the literature, several ILs have been utilised solely for the recovery of both valuable and hazardous metals from DPCBs. For the first time, the research looked into the process of leaching Cu and other important metals from DPCBs using a Brønsted acidic IL known as 1-butyl-3-methyl-imidazolium hydrogen sulfate, [BMIM][HSO_4] [43]. Interestingly, under the optimal conditions, the recovery of Cu surpassed 99.0%, approaching near-total recovery at almost 100.0%. So, after this pioneering work, the use of ILs in the recovery of metals from DPCBs has increased tremendously. For example, a study by Chen's research group examined the leaching characteristics of Cu and Pb found in DPCBs by employing various acidic ILs as leaching agents [44]. In this case, after optimisation, a remarkable Cu leaching performance of almost 100.0% was achieved. However, Pb showed a lower leaching rate (<30.0%). Similarly, under specific optimal conditions involving a 30.0% concentration of H_2O_2, a solid-to-IL ratio (S/IL) of 1:20, a temperature of 80.0 °C, and a duration of 2.0 h, a study conducted by Zhang et al. successfully leached 98.3% of Cu using an acidic IL called 1-carboxymethyl-3-methylimidazolium hydrogen sulfate, [CM-MIM][HSO_4] [45]. Also, the use of sulfonic acid-functionalized IL, N-sulfobutylpyridinium hydrosulfate, [BSO_3HPy][OTf] has been reported and it resulted in leaching efficiency exceeding 99.7% Cu and 74.8% for Zn, albeit with low selectivity for Pb [46].

Barrueto et al.'s research demonstrated selective leaching of different valuable metals which can be influenced by the type of ILs used [47]. Acidic ILs such as [BMIM][HSO_4] and 1-H-3-methyl-imidazolium hydrogen sulfate ([HMIM][HSO_4]) were effective at leaching Cu and Co. On the other hand, basic ILs like 1-butyl-3-methyl-imidazolium bromide ([BMIM][Br]) and 1-butyl-3-methyl-imidazolium chloride ([BMIM][Cl]) were adept at leaching Au and Ag. Consequently, a multi-stage leaching approach can be employed to selectively extract specific metals from DPCBs. The authors observed that [BMIM]HSO_4] acidic IL displayed the best recovery efficiencies for Cu (86.2%) and Co (96.7%), while [BMIM][Br] basic IL displayed the best recovery efficiencies for Au (40.8%) and Ag (44.7%). Similarly, under optimised conditions consisting of 30.0% [BMIM][HSO_4], 10.0% H_2O_2, and 60.0% H_2O at a temperature of 60.0 °C, Gomez et al. recently successfully extracted more than 99.0% of Cu from pre-treated DPCBs [19]. This extraction process was conducted with a S/L

Table 1 Summary of the recovery of metals from DPCBs using ILs, DESs and organic acids at different experimental conditions

Leaching solvents	Concentration of the base leaching solvent	Oxidant	S/L	Particle size	T (°C)	t (h)	Stirring speed (rpm)	Metal recovery (%)	References
[BMIM][HSO_4] & H_2O_2	80.00% IL	30.00% H_2O_2	1.00/25.00	0.10–0.25 mm	70.00	2.00	250.00	≈100.00 (Cu)	[43]
[CM-MIM][HSO_4] & H_2O_2	90.00% IL	30.00% H_2O_2	1.00/20.00	>0.50 mm	80.00	2.00	–	98.30 (Cu)	[45]
[BSO_3HPy][OTf]	80.00% IL	25.00% H_2O_2	1.00/20.00	0.10–0.25 mm	50.00	2.00	250.00	99.70 (Cu) 74.80 (Zn) <10.00 (Pb)	[46]
[BMIM]HSO_4], [BMIM][Br] & H_2O_2	60.00% IL	30.00% H_2O_2	1.00/15.00	2.00 × 2.00 cm	60.00	24.00	50.00	86.20 (Cu) 96.70 (Co) 40.80 (Au) 44.70 (Ag)	[47]
[BMIM][HSO_4] & H_2O_2	30.00% IL	10.00% H_2O_2	1.00/15.00	1.00 × 1.00 cm^2	60.00	2.00	150.00	>99.00 (Cu)	[19]
[NNDMOA][HSO_4] & H_2O_2	0.50 M IL	–	1.00/20.00	1.00 × 1.00 cm	75.00	3.00	500.00	550.00 mg (Cu)	[48]
ChCl:EG (1:2) & I_2	–	0.10 M I_2	–	1.00 × 1.00 cm^2	85.00	72.00	150.00	>75.00 (Cu, Ni, & Sn) ≈45.00 (Zn) <40.00 (Fe) <10.00 (Al, Mg, & Pb)	[59]
ChCl:MA (1:1) H_2O_2 & DDACl	–	30.00% H_2O_2	1.00/10.00	–	60.00	2.00	500.00	8.80 (Cu) 26.40 (Ag) 91.50 (Al) 19.70 (Fe) 2.30 (Zn)	[56]
ChCl:OA (1:2) & ChCl:GA (1:2)	–	1 M OxA & 1 g of Fe	–	–	50.00	8.00	100.00	85.21(Cu) 75.3 (Fe) 83.1(Zn) 22.1 (Sn) 38.7 (Pb)	[57]
CA & H_2O_2	1.00 M CA	5.83% H_2O_2	–	4.00 × 4.00 cm	30.00	4.00	150.00	100.00 (Cu, Sn, Zn, Ni, Au, Pb, Al, Fe, Ag, & Pd)	[61]
CA & H_2O_2	0.50 M CA	5.83% H_2O_2	–	7.00 × 7.00 mm	30.00	<4.00	150.00	85.00 (Cu) 98.00 (Al) 94.00 (Pb)	[62]
CA, IA & OxA	50.00 mM CA, 30.00 mM IA & 20.00 mM OxA	–	–	60.00–80.00 mesh	–	15.00 days	–	80.3 (Cu) 75.70 (Zn) 73.60 (Ni)	[60]
CA, AA & H_2O_2	1.00 M CA, 5.00% AA & 5.00% H_2O_2	30.00% H_2O_2	–	3.00 × 3.00 cm	30.00	24.00	–	325.89 ppm (Cu)	[63]
Lemon juice & H_2O_2	74.00% Lemon juice	12.20% H_2O_2	1.41%	149.00–177.00 µm	20.00	4.00	200.00	89.00 (Cu) 73.00 (Zn)	[64]

Table 1 (continued)

Leaching solvents	Concentration of the base leaching solvent	Oxidant	S/L	Particle size	T (°C)	t (h)	Stirring speed (rpm)	Metal recovery (%)	References
MSA & H_2O_2	3.50 M MSA	0.50 M H_2O_2	–	7.00 × 7.00 mm	30.00	1.50	150.00	35.98 ppm (Cu) 1254.00 ppm (Pb) 822.30 ppm (Sn)	[62]
MSA & H_2O_2	1.00 M MSA	0.60 M H_2O_2	1:20	2.00 × 2.00 cm	50.00	2.00	500.00	100.00 (Cu) 100.00 (Zn) 90.00 (Ni)	[70]

ratio of 1:15 for a duration of 2 h. The recovered Cu was then obtained through a direct electrowinning process using the [BMIM][HSO_4]-leach solution as the electrolyte. This work employed minimal leaching agents, time, and energy compared to the previous works. Furthermore, the combination of N,N-dimethyloctylammonium hydrogen sulfate ([NNDMOA][HSO_4]) and H_2O_2 proved effective in leaching 550.00 mg of Cu from DPCBs [48]. However, this particular IL did not demonstrate successful leaching of significant amounts of Au and Ag. This limitation can be attributed to the preference of Au and Ag for a basic environment over an acidic one [47].

In the process of recovering metals from DPCBs, conventional slurry electrolysis demands substantial amounts of strong acids or strong alkalis to form the electrolyte solution, leading to the discharge of pollutant waste acids or alkalis. Moreover, traditional slurry electrolysis involves complex operational processes and stringent conditions. Consequently, there is a pressing need to develop environmentally friendly electrolysis technologies for metal recovery that offer simplicity in operation, reduced reagent consumption, and minimal pollutant emissions [49].

The recovery of metals from DPCBs has advanced to the point whereby ILs are being employed in slurry electrolytic processes. The schematic diagram for this process is shown in Fig. 2(a). For example, N-butyl sulfonate pyridinium bisulfate ([BSO_3HPy]HSO_4]) IL, was used to replace H_2SO_4 in a slurry electrolytic system for one-step Cu recovery from DPCBs [50]. The findings reveal that substituting H_2SO_4 with [BSO_3HPy]HSO_4] can enhance Cu recovery rate, current efficiency, and purity. The optimal outcome is achieved when 10.0% H_2SO_4 is replaced, resulting in a 90.9% recovery, 70.7% current efficiency, 81.7% purity and Cu powder particle size of 2.3 µm. In a similar context of slurry electrolysis, employing an IL solution containing $CuSO_4$, NaCl, H_2SO_4, and 1-butyl-3-methylimidazolium hexafluorophosphate, [BMIM][PF_6] as reported by He et al. in 2020, demonstrated improved performance with a 92.7% recovery rate and 80.0% current efficiency for Cu [49]. Moreover, 1-ethyl-3-methylimidazole chloride, [EMIM][Cl] and1-butyl-3-methylimidazole hexafluoroborate, [BMIM][BF_6] were identified as ideal ILs for achieving high recoveries of Cu.

The most recent work on the recovery of metals from DPCBs is the modification of adsorbent with ILs for the sole aim of enhancing their capacity. For instance, Lin et al. developed an eco-friendly adsorbent using 1-(3-aminopropyl) imidazole (APIM) IL functionalised with chitosan fibres (CFs) to selectively recover Au from DPCBs bioleachate [51]. The APIM-CFs were created for Au (I) recovery under alkaline conditions, with uniform cross-sectional diameter. IL-CFs had a maximum Au adsorption capacity of 357.0 ± 13.2 mg/g, reaching equilibrium in 7.0 min. Pseudo-1st-order and pseudo-2nd-order rate constants were 0.89 ± 0.061/min and 0.0049 ± 0.0002 g/mg min. APIM-CFs exhibited high Au selectivity from DPCBs bioleachate, suggesting their effectiveness in recovering Au from urban mining bioleachates. The merits and demerits of employing ILs in the recovery of metals from DPCBs compared to conventional methods are highlighted in Fig. 2(b).

Recovery of metals mediated by deep eutectic solvents

In this subsection, we reviewed research publications that dealt with the leaching of metals from DPCBs using environmentally friendly solvents popularly known as DESs. In recent years, the use of DESs in separation technology has generated increasing research interest. DESs are mixtures of two or more compounds that form a liquid phase at a lower temperature than the melting point of each compound [52–54]. In this case, one component of the DES is known as a hydrogen bond donor (HBA) and the other is normally referred to as a hydrogen bond

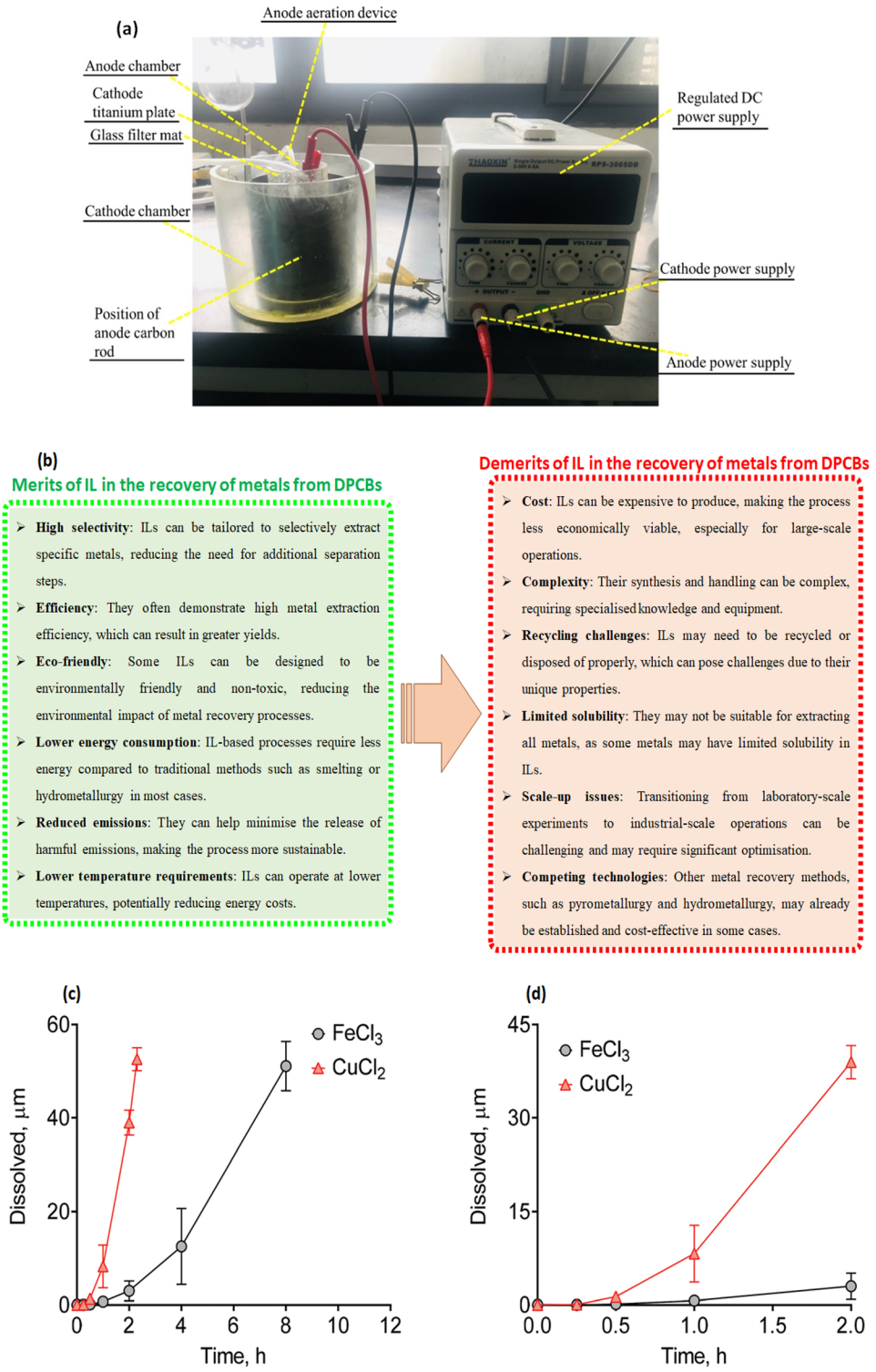
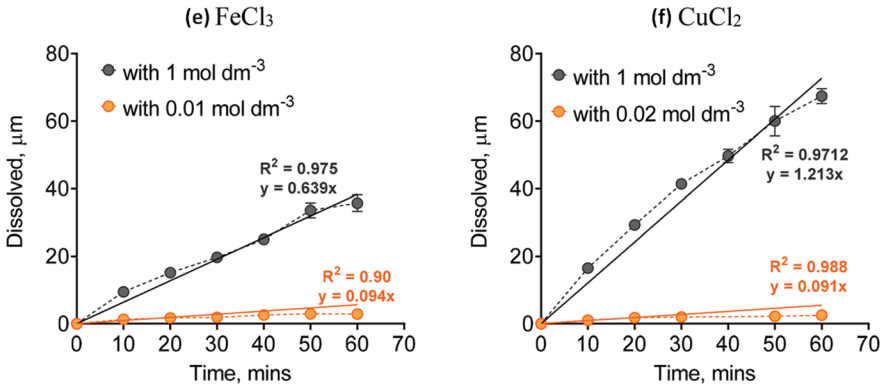


Fig. 2 **a** Experimental setup for the electrolytic slurry process, reprinted from [49], Copyright 2020, with permission from Elsevier, **b** Merits and demerits of using ILs for the recovery of metals from DPCBs, **c** Etch depth of a Au-coated PCB after oxidation in $CaCl_2{\cdot}6H_2O$:EG DES with either 1 M $FeCl_3$ or $CuCl_2$, at 50 °C, **d** Etch depth in the first 2.0 h of etching; etch depth of Cu over time from the cross-section of a square-shaped block terminal with two different concentrations of **e** FeCl3 and **f** $CuCl_2$ as oxidising agents in the $CaCl_2{\cdot}6H_2O$:EG eutectic system at 50 °C. (**c–f**) was reproduced from [58], Copyright 2022, with permission from RSC

acceptor (HBD). DESs have several advantages over conventional solvents, such as low toxicity, low cost, high biodegradability, and high solubility of metals [22, 55]. DESs can be used to leach metals from DPCBs by dissolving the metal oxides or salts present on the surface of the boards. The leached metals can then be recovered by various techniques, such as precipitation, electroplating, or solvent extraction. Only a few research papers have reported the leaching of metals from DPCBs using DESs [56–59].

Fig. 2 (continued)



The pioneering work involving the use of DESs in the leaching of metals from DPCBs was reported in 2021 by Kolliopoulos' research team [59]. In this study, the researchers focused on extracting metals from DPCBs using choline chloride (ChCl) and ethylene glycol (EG) DES. Specifically, they successfully extracted Cu, Ni, Zn, and Sn among others from the DPCBs. The leaching efficiency was impressive, with over 75.0% recovery achieved after 72.0 h for Cu, Ni, and Sn, and approximately 45.0% recovery for Zn when I_2 was employed as oxidizing agent. In the absence of I_2 in ChCl:EG DES, the recovery of metals was generally observed to be low. In that case, less than 15.0% recovery was observed for most metals, except Sn, where leaching was about 100.0%.

Recently, the recovery of metals from DPCBs using DES has also been carried out by Abbott's research group [58]. In this study, researchers confirmed that $FeCl_3$ and $CuCl_2$ have high solubility in $CaCl_2{\cdot}6H_2O$:EG DES and exhibit reversible redox behaviour. These properties make these inorganic salts suitable as strong and electrocatalytic oxidising agents. They used optical profilometry to measure the dissolution rates of Cu, Ni, and Au in a PCB block terminal. Cu showed the fastest dissolution rate, while Ni did not dissolve, likely due to the high water content in the eutectic mixture. $CuCl_2$ was found to be a more effective oxidizing agent than $FeCl_3$, requiring less time for etching. In other words, their study explores the use of catalytic dissolution in a eutectic solvent for selectively recovering metals from DPCB. $FeCl_3$ and $CuCl_2$ serve as effective oxidising agents, with $CuCl_2$ demonstrating faster leaching kinetics compared to $FeCl_3$ as illustrated in Fig. 2 **(c–f)**.

Furthermore, the possibility of extracting and separating metals from DPCBs, either through leaching or without the need for high-temperature acidic leaching was investigated using two commonly employed DESs: {ChCl + lactic acid [LA], 1:2} and {ChCl + malonic acid [MA], 1:1} [56]. The authors added H_2O_2 as an oxidant and didecyldimethylammonium chloride (DDACl) as a surfactant primarily to improve the leaching efficiency of the DESs utilised. Interestingly, much higher leaching efficiency was obtained for Al (91.5%) and other metals at pH 2.5 when ChCl:MA DES was used compared to when ChCl:LA DES was employed.

Most recently, three different DESs were prepared and used in a two-step process for the recovery of various metals from DPCBs [57]. In this study, ChCl served as HBA, while EG, oleic acid (OA), and glycolic acid (GA) were employed as HBDs in separate eutectic mixtures. Zn, Cu, and Sn recovery was exclusively accomplished through the introduction of oxalic acid (OxA), water, and Fe powder into DESs. In other words, when oxalic acid solution was added to the ChCl:GA DES, a blue precipitate formed. This precipitate comprised $ZnC_2O_4{\cdot}2H_2O$, $PbC_2O_4{\cdot}2H_2O$, and $CuC_2O_4{\cdot}2H_2O$, with recovery yields of 90.4% Zn, 87.5% Pb, and 16.8% Cu. The residue was subjected to ChCl:OA DES leaching. In this case, Cu was successfully recovered by mixing the ChCl:OA DES leachate with water. Specifically, 74.9% of Cu was precipitated as $CuC_2O_4{\cdot}2H_2O$ with a purity of 98.1 wt%. The remaining Sn in the aqueous solution was efficiently recovered by adding reduced Fe powder, resulting in a Sn recovery yield of 51.3%. Nevertheless, it is possible to enhance the recovery yields of the desired metals and improve the purity of $CuC_2O_4{\cdot}2H_2O$ by fine-tuning the experimental conditions.

Research into the utilisation of DESs for recovering metals from DPCBs is relatively scarce as mentioned earlier. This scarcity of research could be attributed to the greater complexity in the compositions of DPCBs sourced from various origins when compared to the other simpler waste types. Nevertheless, the solvometallurgical approach involving the use of DESs for the recovery of metals from DPCBs offers a promising alternative that reduces the need for excessive water and acidic/basic reagents in the recovery process. The results obtained so far by different research groups demonstrate its potential to compete with existing methods of metal leaching, pointing towards the achievement of a sustainable circular economy through zero-waste green urban mining.

Recovery of metals mediated by organic acids

An alternative approach to the use of harmful inorganic acids during the recovery of metals from DPCBs also entails utilising natural acids such as citric acid (CA), itaconic acid (IA), and oxalic acid (OxA), among others. These solvents are biodegradable, less harmful, and exhibit greater selectivity compared to their inorganic counterparts. Organic acids can be produced by microorganisms, such as fungi or bacteria, through fermentation processes [60]. This renders organic acid leaching a more environmentally friendly and sustainable method for extracting metals from DPCBs.

Interestingly, numerous research studies have investigated the utilisation of organic acids in metal recovery from DPCBs. CA stands as an eco-friendly and auspicious leaching substance for extracting valuable metals from PCBs. For instance, Jadhav et al. in their recent study completely dissolved a range of metals, including Cu, Sn, Zn, Ni, Au, Pb, Al, Fe, Ag, and Pd using CA (1.0 M) and 5.8% H_2O_2 [61]. Furthermore, CA readily dissolves in water and can be readily broken down in both aerobic and anaerobic environments just like most organic acids. Also, Anwer et al. devised a systematic procedure for separating components and extracting metals from DPCBs using both organic and inorganic acids [62]. They discovered that CA could serve as an eco-friendly substitute for inorganic acids, yielding comparable or superior results as presented in Table 1. The outstanding ability demonstrated by the solution of CA and H_2O_2 during the recovery of metals from DPCBs can be attributed to strong oxidant formation (i.e., peroxyl carboxylic acid) during the leaching process. Similar to H_2O_2, these substances readily undergo decomposition in an aqueous environment, leading to the formation of the peroxy carboxyl group (–COOOH), which in turn oxidizes the metal. The reactions that took place during the leaching process which led to the formation of peroxyl carboxylic acid are shown in Eqs. (1 & 2) [61].

$$R{-}COOH + H_2O_2 \rightarrow R{-}COOOH - H_2O \quad (1)$$

$$R{-}COOOH + M + 2H^+ \rightarrow R{-}COOH + H_2O + M^{2+} \quad (2)$$

In the above equations, "R" represents the organic substituent group, and "M" represents the metal. According to the reaction shown in Eq. (3), there is a potential alternative pathway for the generation of metal hydrogen citrate when both H_2O_2 and CA are present [62].

$$C_6H_8O_7(CA) + M + H_2O_2 \rightarrow M(C_6H_8O_7) \text{ i.e. metal hydrogen citrate} + H_2O + M^{2+} \quad (3)$$

In addition, Krishnamoorthy et al. harnessed organic acids (i.e., a mixture of 50.0 mM CA + 30.0 mM IA + 20.0 mM OxA) produced by *Aspergillus niveus* for the leaching of various metals from DPCBs [60]. The authors achieved a maximum recovery of 75.7% for Zn, 73.6% for Ni, and 80.3% for Cu from DPCBs using a two-step leaching process with particle sizes ranging from 60.0 to 80.0 mesh. Recently, Nagarajan and Panchatcharam explored the combination of CA and acetic acid (AA) solution for Cu leaching from DPCBs in the presence of H_2O_2 [63]. Their observations revealed that when CA, AA, and H_2O_2 were applied individually for the leaching process, lower Cu concentrations were achieved compared to using a leaching solution composed of 1.0 M CA, 5.0% AA, and 5.0% H_2O_2. In specific terms, leaching with CA, AA, and H_2O_2 yielded Cu concentrations of 26.9, 22.3, and 6.3 ppm, whereas the combined solution of these three chemicals resulted in a Cu concentration of 325.9 ppm. Therefore, the combination of organic acids could be established as a standardised method for leaching Cu and other metals present in DPCBs.

Moreover, natural organic acids obtained from agricultural fruits have also been utilised for the leaching of metals from DPCBs and are found to be effective. One such work is the one recently carried out by Rastegar's research team [64]. In their study, they obtained natural acid from lemon juice, and this was used in combination with H_2O_2 to recover Cu and Zn from DPCBs successfully. The authors applied the response surface method to optimise the extraction which resulted in the maximum recoveries of 89.0 and 73.0%, respectively, for Cu and Zn.

The use of methanesulfonic acid (MSA) is trending currently as a leaching agent for metal recovery from DPCBs. MSA is a biodegradable and non-volatile organic acid that has been widely used in industrial applications, such as electroplating, metal cleaning, and organic synthesis [65, 66]. The minimal toxicity of MSA becomes apparent when examining its lethal dose 50 (LD_{50}) value. The reported LD_{50} (oral, cat) for MSA is 1158 mg/kg, in stark contrast to the LD_{50} (oral, rat) of HCl, which falls within the range of 238–277 mg/kg [67]. The chemical makeup of MSA can be likened to that of H_2SO_4, with the exception that one of the two hydroxyl (OH) groups has been replaced by a methyl (CH_3) group. MSA demonstrates remarkable acidity, as indicated by its low pKa value of −1.19 [68]. Consequently, MSA is significantly more acidic when compared to other organic acids such as formic acid, AA, or CA. Its level of acidity rivals that of potent mineral acids. Additionally, in a solution with a concentration of 0.1 M, MSA undergoes almost complete ionisation. The heat released during the formation of the methanesulfonate ion in an aqueous solution is −163.79 ± 1.04 kcal/mol, and the heat of formation for liquid MSA is −152.39 ± 1.12 kcal/mol at 25 °C [69].

In summary, MSA has several advantages over conventional acids like H_2SO_4 or HCl in metal leaching.

So, MSA can selectively dissolve the solder (Sn–Pb alloy) from the DPCBs without affecting the epoxy resin or the Cu interconnects [62]. The reaction for this process is shown in Eq. (4). This facilitates the separation of the metallic and non-metallic components of the DPCBs and reduces the consumption of acid and energy. MSA can leach Cu and other base metals from the DPCBs at low concentrations and temperatures. For example, a study by Anwer et al. showed that 3.5 M MSA and 0.5 M H_2O_2 can dissolve Cu, Pb and Sn from DPCBs in 1.5 h at 28.0 °C 62.

$$2\text{MSA}-\text{H}_2\text{O}_2 + \text{Sn} + \text{Pb} + 4\text{H}^+ \rightarrow \text{Sn}^{2+} + \text{Pb}^{2+} + 2\text{MSA} + 4\text{H}_2\text{O} \quad (4)$$

Furthermore, Jadhao et al. investigated the recovery of base metals like Cu, Zn, and Ni from DPCBs using MSA and H_2O_2 [70]. After optimisation, the authors were able to successfully leach 100.0% Cu and Zn in addition to 90.0% Ni. They also found that the extraction of these metals is primarily controlled by diffusion in which the activation energies for Cu, Zn, and Ni extraction were estimated to be 9.4, 10.9, and 18.9 kJ/mol, respectively. By combining electrowinning and cementation, 99.9% purity was obtained for Cu and Zn. A typical overall method for recovering metals from DPCBs using MSA is depicted in Fig. 3.

In summary, the use of organic acids, whether in isolation or as mixtures, coupled with optimisation techniques, presents a promising avenue for eco-conscious metal recovery from DPCBs. Additionally, the adoption of MSA, with its selective solder dissolution and low toxicity, shows great promise for reducing environmental harm and energy consumption while recovering base and precious metals from DPCBs. These innovative approaches can contribute to the sustainable management of e-waste and underscore the importance of eco-friendly alternatives in the field of metal recycling.

Recovery of metals mediated by amino acids

Proteins are made of organic molecules called amino acids. Amino acids have raised significant attention in research across various scientific domains within the fields of chemistry and biology. There are 20 common

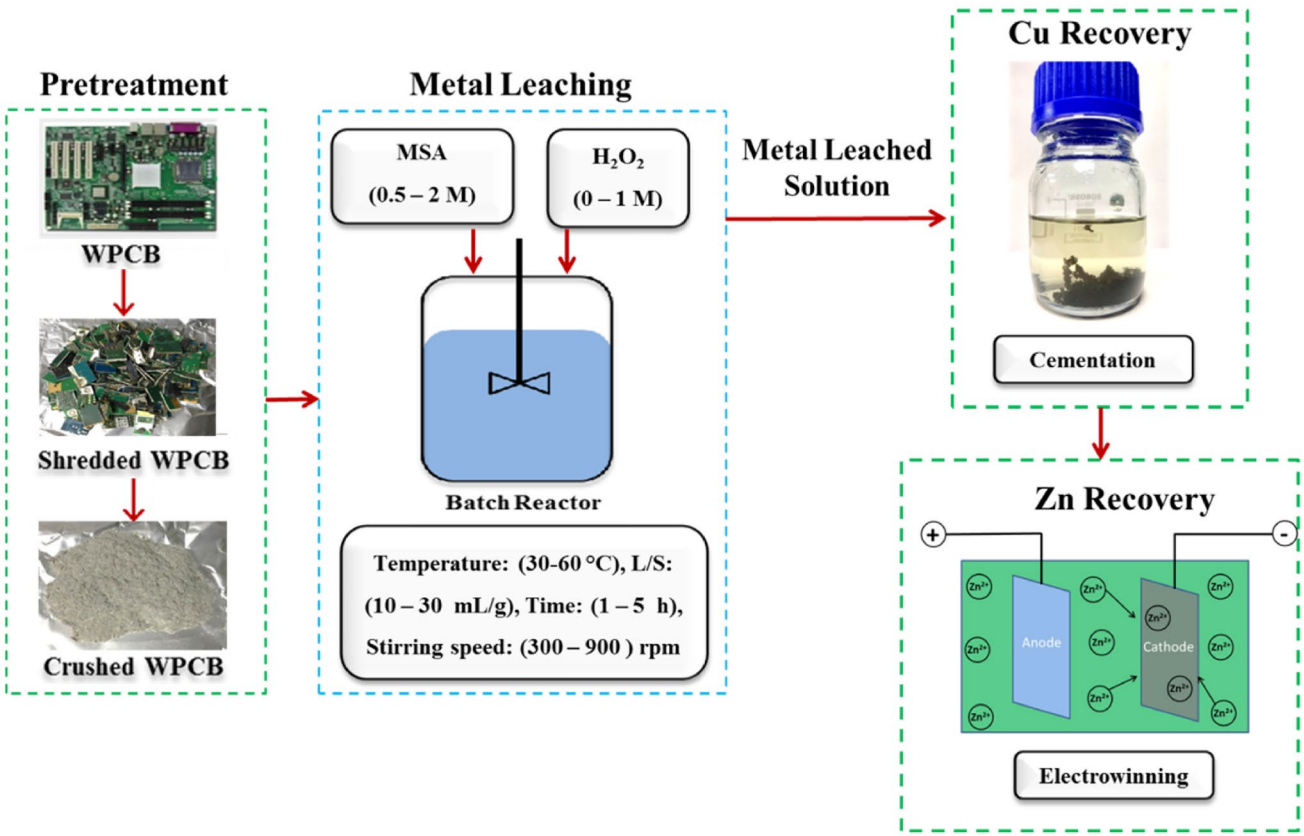


Fig. 3 A typical overall recovery methodology of metals from DPCBs using organic acids, reproduced from [70], Copyright 2023, with permission from Elsevier

types of amino acids, known as α-amino acids, that have a carbon atom (α-carbon) with three groups attached to it: a carboxyl group (–COOH), an amino group ($–NH_2$), and a variable group (called the R-group) [71]. These amino acids are weak acids that can lose more than one hydrogen atom in water. The amount of hydrogen atoms that are lost depends on the pH of water [72]. The simplest amino acid is glycine (NH_2CH_2COOH), and it is the most widely used amino acid for the recovery of metals from DPCBs. Amino acids are usually employed as lixiviants in an alkaline setting to extract base and precious metals from DPCBs. NH_2CH_2COOH is industrially produced. It is cost-effective and possesses attributes such as non-toxicity, non-volatility, and non-combustibility [73]. In numerous research studies, NH_2CH_2COOH has been favoured among amino acids due to its lower cost and abundant availability [74]. It has demonstrated the ability to reduce the requirement for high cyanide concentrations in NH_2CH_2COOH-cyanide solutions [75]. Furthermore, NH_2CH_2COOH can dissolve Au independently, without the need for cyanide [76]. When common oxidants are introduced to alkaline-NH_2CH_2COOH solutions, it was observed that they can enhance the rate and efficiency of Au leaching [77]. Therefore, in this section, we elaborated on the recovery of metals from DPCBs using amino acids, most especially NH_2CH_2COOH as a lixiviant.

Fascinatingly, many studies have explored the recovery of Cu and other metals from DPCBs by employing NH_2CH_2COOH as a lexiviant. It is worth noting that NH_2CH_2COOH tends to form complexes with Zn and Pb more readily than with precious metals [78]. This behaviour primarily arises from the standard reduction potential of base metals, which is typically higher than that of precious metals. This makes the oxidation of base metals more straightforward. For example, Li et al. achieved a maximum Cu recovery of 96.5% (the reaction mechanisms are shown in Eqs. 5 and 6), but this process resulted in significant co-extraction of base metals [79]. The concentration of NH_2CH_2COOH and the solid content had a notable impact on the extraction of base metals, while factors like H_2O_2, temperature, and particle size had minimal influence. Their kinetic study, conducted at room temperature with ambient O_2 as the oxidant and particle sizes below 2 mm, suggested that the extraction of Cu from DPCBs is governed by internal diffusion. For example, Han et al. also investigated the effective parameters for Cu leaching from DPCBs using NH_2CH_2COOH [80]. Interestingly, their findings indicated that increasing the H_2O_2 content improved Cu leaching efficiency. In a similar study, Li et al. demonstrated that Cu could be efficiently precipitated as CuS with an optimised Cu/HS ratio of 1:1.2, achieving over 99.0% precipitation efficiency in less than 5 min [81]. Most recently, a complex four-stage process of Cu leaching from PCB waste powder using NH_2CH_2COOH was studied by Mokhlis et al. under ambient conditions [82]. During this process, NH_2CH_2COOH displays remarkable selectivity for Cu, enabling the dissolution of Cu from DPCB waste at a substantial rate and 92.8% recovery.

$$2Cu + O_2 \rightarrow 2CuO \tag{5}$$

$$CuO + H_2O + 2NH_2CH_2COO^- + \rightarrow Cu(NH_2CH_2COO_2) + 2OH^- \tag{6}$$

Furthermore, Broeksma and Dorfling studied the optimal conditions for achieving the highest Cu dissolution and the lowest Ag dissolution recently from DPCBs [83]. They found that a temperature of 60.0 °C, a NH_2CH_2COOH concentration of 1.0 M, the utilisation of O_2 as the oxidant, a pH level of 11.0, and a pulp density of 25.0 g/L are required. Under these conditions, a remarkable 81.2% Cu dissolution was achieved within 22.0 h, accompanied by minimal co-extraction of Au, which accounted for just 1.3%. A system composed of $NH_2CH_2COOH + H_2O_2$ proved effective for extracting Cu and base metals, with NH_2CH_2COOH concentration being the most influential factor, followed by the H_2O_2 concentration [84]. Higher pulp density negatively impacted the Cu leaching efficiency. Under optimal conditions (0.5 M NH_2CH_2COOH, 1.0% H_2O_2, and 20 g/L pulp density) at ambient temperature, 99.9% of Cu was extracted, and 95.2% of it precipitated as CuS. Also, Ag recovery from the residue was achieved using the $NH_2CH_2COOH + KMnO_4$ system, with 96.0% recovery at ambient temperature under optimal conditions.

Monosodium glutamate ($C_5H_8NO_4Na$), derived from glutamic acid and known as one of the most abundant non-essential amino acids in nature, has also recently found application in the recovery of metals from DPCBs. For instance, the leaching of Cu and Au from DPCBs was achieved recently under pH 7.0 (neutral pH) using $C_5H_8NO_4Na$ as the base lixiviant [85]. The highest Cu and Au extractions were achieved through a two-stage leaching process conducted under mild conditions (30.0 °C, 150.0 rpm, and an initial pH of 7.0). In the initial leaching stage, a significant 93.0% of Cu was dissolved with minimal contamination of other metals, using a combination of 0.75 M $C_5H_8NO_4Na$ and 0.5% H_2O_2 (as shown in Eqs. 7 & 8). In the subsequent leaching stage, Au (as presented in Eq. 9) extraction was maximised at 86.0% from the solid residue by employing 1.0 M $C_5H_8NO_4Na$ supplemented with 0.25% H_2O_2. Following this, a direct electrowinning process enabled the recovery of 98.0–100.0% of Cu from the initial leachate within 5.0–6.0 h.

Additionally, Perea et al. explored how the choice and concentration of oxidising agents affect Cu leaching kinetics. They found that using 0.5 M $C_5H_8NO_4Na$ and 0.03 M H_2O_2 resulted in the highest Cu extraction of 92.0% after 2.0 h.

Moreover, $C_5H_8NO_4Na$ outperformed NH_2CH_2COOH in both dissolution kinetics and extraction rate, with maximum recovery values of 92.0 and 84.0%, respectively.

$$Cu^{2+} + C_5H_8NO_4Na \rightarrow Cu(C_5H_8NO_4)^+ + Na^+ \quad (7)$$

$$Cu^{2+} + 2C_5H_8NO_4Na \rightarrow Cu(C_5H_8NO_4)_2 + 2Na^+ \quad (8)$$

$$Au^{2+} + 2C_5H_8NO_4Na \rightarrow Au(C_5H_8NO_4)_2^- + 2Na^+ \quad (9)$$

Furthermore, a few of the recent studies deal with an integrated approach involving the use of amino acids and other substances. For example, in a recent study conducted by Li et al., an innovative approach was adopted to enhance both the solid content and the leaching efficiency in the NH_2CH_2COOH-oxidant system [86]. In this study, NH_3, a well-known Cu lixiviant, was introduced into the process. It was discovered that NH_3 served multiple roles as a pH modifier, an additional lixiviant, and a potential catalyst during the NH_2CH_2COOH-based leaching of DPCBs. This modification resulted in a significant reduction in leaching time, from 72.0 to 24.0 h, and an increase in solid content to as high as 15.0%, yielding more than 40.0 g/L of Cu in the pregnant leach solution. However, it is essential to implement additional safety precautions to mitigate the potential hazards associated with NH_3, despite its reduced usage by 40.0–80.0% compared to traditional NH_3–NH_4 leaching methods [87]. Also, in another study by the same research group, NH_2CH_2COOH was employed alongside oxidants, either $KMnO_4$ or potassium ferricyanide ($K_3[Fe(CN)_6]$), to extract precious metals, Au, Ag, and Pd, from the residue remaining after the initial NH_2CH_2COOH leaching of DPCBs [88]. Under optimised conditions, which include 0.5 M NH_2CH_2COOH, room temperature, a pH level of 11.0, and 2.0% solids content, the NH_2CH_2COOH-$KMnO_4$ combination yielded higher extractions of these metals. These extraction efficiencies were found to be comparable to cyanide-based leaching methods. However, it is worth noting that the consumption of oxidants was relatively high, with 632.0 kg/t of $KMnO_4$ and 610.0 kg/t $K_3[Fe(CN)_6]$ used under the optimised conditions. Owing to this, further research is recommended to explore oxidant reduction or regeneration methods and to investigate leaching at higher solids content, such as levels exceeding 10.0%, which would complement the findings of this study.

Recently, a novel leaching system was developed by utilising thiosulfate-cobalt-glycine for Au recovery from PCB of mobile phones [89]. This innovative system effectively addresses the challenge of high thiosulfate consumption that limits the commercial use of conventional thiosulfate leaching. Notably, the authors achieved an impressive 97.8% Au recovery using the thiosulfate-cobalt-glycine system. Furthermore, the research investigated key parameters, revealing that higher concentrations of Co(II), thiosulfate, or glycine enhance Au leaching, while elevated temperatures can lead to thiosulfate decomposition and reduced Co stability, which negatively impacts Au leaching efficiency. A similar study conducted by Godigamuwa and Okibe highlighted the growing significance of thiosulfate and glycine systems for Au leaching from DPCBs [90]. The authors found that the glycine-thiosulfate system outperformed the histidine-thiosulfate system, achieving an impressive Au leaching efficiency of 93.7% at a pH of 9.3 and a temperature of 40 °C. In comparison, the stand-alone thiosulfate and glycine systems achieved Au leaching percentages of 47.1 and 50.7%, respectively. Notably, in the dual system, Fe leaching was insignificant, interestingly, the Ag and Al leaching reached 95.3 and 27.0%, respectively. Furthermore, when compared to the thiosulfate system, the dual system demonstrated enhanced stability in leached Au. The optimal conditions for Au leaching from PCBs in the dual system were found to be 60.0 mM thiosulfate and 0.5 M glycine at 40 °C and a pH of 9.3. Kinetic analysis revealed that Au and Ag leaching in the dual system followed a diffusion-controlled model. Interestingly, the initial Au leaching rate in the dual system was observed to resemble that of the glycine-cyanide system.

In most cases, using amino acids on their own cannot achieve a satisfactory extraction of metals. Additives, including oxidants/reductants, catalysts, synergists, pH modifiers, etc., are often used alongside amino acids, to mobilise metals from different material matrixes to leaching solution. Figure 4(a) illustrates a typical amino acid leaching system used in hydrometallurgy. Oxidants or reductants are usually used to oxidise or reduce metal ions to their preferred oxidation state before complexing with amino acids. For example, when leaching DPCBs, where Cu presents as its native form, oxidant (ambient O_2/H_2O_2 etc.) is indispensable in the system to oxide metallic Cu to cupric form that is to be dominantly complexed with amino acid [79]. The selection of the appropriate redox agent is important and system-dependent and care should be taken to not destroy the amino acid itself in the process of amino acid recycling and reuse. Catalysts and synergists are used to catalyse the reaction of amino acids with metals and to be synergistic with amino acids during the leaching process. A typical example is the use of starved cyanide in the amino acid leaching of Ag. Several studies have shown that the addition of starved levels of cyanide (no "free" cyanide in the reactor exit) to the amino acid solution could enhance the leaching kinetics of Ag significantly [86–88]. Additionally, as indicated in Fig. 4(b), different amino acid species will predominate at different pH of the leaching system. A pH modifier is hence required for an amino acid leaching system, such as NaOH, CaO, ammonia water ($NH_3 \cdot H_2O$) and dilute mineral acids, which

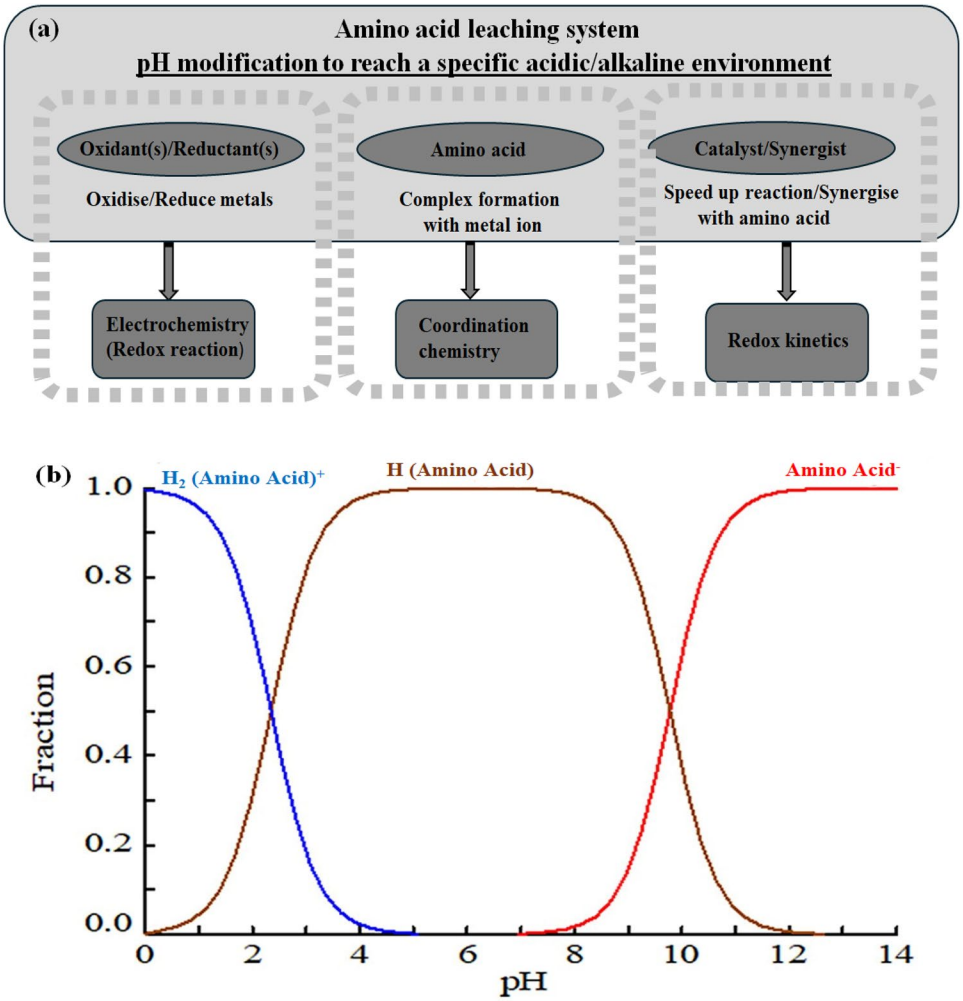


Fig. 4 **a** Basic illustration of the hydrometallurgical system employing amino acid leaching, **b** Fraction of amino acid species in water at 25 °C and different pH, adapted from [87], Copyright 2023, with permission from Elsevier

is to make the preferred amino acid species predominant in the system for effective coordinative reaction.

When dealing with the leaching of metals from DPCBs using amino acids or an amino acid-integrated approach, it is advisable to carry out leaching in two distinct stages. During the first stage, base metals, comprising primarily Cu and Zn (over 90.0%), can be efficiently extracted at room temperature and under normal environmental conditions. However, the second stage of leaching, which targets precious metals, necessitates the introduction of an additional oxidant, such as H_2O_2, and/or a synergist like starved cyanide. This stage also covers the extraction of the remaining base metals. It is important to note that because some metals are embedded within the polymer substrate of DPCBs, fine grinding or pulverising of materials could be required for the successful leaching of precious metals and the remaining base metals. This additional step may incur extra costs in the process.

Factors affecting the recovery of metals from DPCBs

The recovery of metals from DPCBs is a complex and highly controlled process, heavily influenced by a multitude of critical factors. These factors encompass a range of physical and chemical parameters, including pH levels, temperature, particle size, leaching agent concentrations, oxidant concentrations, reaction time, and many others. The interplay of these variables can significantly impact the efficiency and yield of metal recovery from DPCBs, making it a precise science that requires careful consideration and optimisation. Therefore, the discussion in this section focuses on the intricacies of these influencing factors and their crucial roles in determining the success of DPCB metal recovery processes. Understanding how pH, temperature, particle size, and other variables affect leaching and extraction processes is essential for developing efficient and sustainable methods for reclaiming valuable metals from e-waste in general. By unravelling the complexities of these factors, researchers can work towards more effective and

environmentally responsible solutions for recycling DPCBs and conserving precious resources.

Effect of contact time and temperature

In general, longer contact times between the solvent and PCBs tend to enhance metal recovery, as it allows for a more complete dissolution of metals from the complex e-waste [19, 45]. However, prolonged exposure may also lead to increased energy consumption and may not always result in a proportional increase in metal yields. Optimising the leaching time is, therefore, crucial, and recent studies emphasise the need to strike a balance between contact time and resource efficiency. Temperature is another critical factor that affects metal recovery from PCBs. Higher temperatures generally accelerate the leaching process by increasing the solubility of metals in the chosen solvents [83]. For example, the leaching rate of Cu from the PCBs increased with time and temperature [19]. After 1 h, the leaching efficiency reached a stable level at different temperatures: 55.0 at 20.0, 66.0 at 40.0, 85.0 at 60.0 and 92.0% at 80.0 °C. However, at 60.0 °C, the leaching efficiency continued to rise until it reached 90.0% in 2.0 h. Thus, it is highly important to strike a balance between the energy consumption required and the yield obtained in the leaching reaction. The effect of contact time and temperature on the leaching efficiency of metals from DPCBs are shown in Fig. 5 (**a** & **b**), respectively. Also, similar observations were noticed for Cu and other metals by other researchers [48, 70]. This is because higher temperature makes the reactant particles move faster and collide more often, which helps them overcome the energy barrier for the reaction. This means that temperature acts like a catalyst and speeds up the reaction [49, 83]. Nevertheless, it is essential to carefully control the temperature to prevent thermal degradation of the solvent or undesirable side reactions.

Effect of concentration of leaching agents

The concentration of leaching agents is one of the important factors that affect the recovery of metals from PCBs. Different leaching agents have different mechanisms and efficiencies for dissolving metals from PCBs. In most cases, the recovery of metals from PCBs tends to increase as the concentration of the leaching agent increases [45, 83]. This is because a higher concentration provides a larger number of reactive ions or molecules, increasing the chances of contact with the target metals. However, leaching agents at higher concentrations after reaching the optimum could decrease the recovery of metals. For example, the work of Nagarajan and Panchatcharam investigated the influence of CA concentration on metal leaching [63]. The leaching process occurred over 24.0 h at 30.0 °C under static conditions, utilising 5.0% H_2O_2 and 5.0% AC. The CA concentration varied from 0.5 to 2.5 M. The researchers found that the most effective CA concentration for Cu leaching is 1.5 M. At this concentration, leaching performance is enhanced because metal-citrate complexes are formed, which are subsequently broken down by H_2O_2 to release free metal ions into the solution. However, when CA concentration surpasses 1.5 M, it leads to the formation of more metal complexes with citrate, but a deficiency of H_2O_2 prevents their conversion into metal ions, resulting in reduced leaching efficiency.

Effect of presence/concentration/nature of oxidants

Oxidants play a pivotal role in the recovery of metals from PCBs during recycling processes. Their primary function is to facilitate the dissolution of metals by promoting oxidation on the PCB surfaces. This oxidation makes the metals more amenable to dissolution by leaching agents, effectively breaking down metal oxides and sulfides [84, 91]. This, in turn, enables the release of valuable metal ions into the leach solution, a critical step in the metal recovery process. Oxidants such as H_2O_2, I_2, $KMnO_4$, ($K_3[Fe(CN)_6]$ or O_2 not only enhance metal dissolution but also accelerate the overall leaching reaction kinetics, reducing the time required for metal extraction [80, 87]. This accelerated reaction speed is particularly advantageous for large-scale recycling operations, improving process efficiency. In the presence of I_2, the recovery rates for Cu, Sn, and Ni exceeded 75.0%, and Zn showed an extraction rate of approximately 45.0% [59]. Conversely, in the absence of I_2 within the ChCl:EG DES solution, metal recovery was generally less than 15.0%, except for Sn, which exhibited an extraction rate of around 100.0% from DPCB [59]. This underscores the vital role played by the presence of I_2 in augmenting the leaching and extraction processes, particularly for metals like Cu, Ni, and Zn, stressing its significance in optimising the recovery of metals from the solution.

Furthermore, as depicted in Fig. 5(c), increasing $KMnO_4$ (oxidant) concentration improved Cu leaching kinetics initially but caused decreased extraction after 3.0 h [92]. This behaviour could be due to overpotential and Cu precipitation [93]. The highest Cu extraction (77.6%) was achieved at 0.01 M $KMnO_4$ after 6.0 h, while both assays reached nearly 72.0% extraction after 24.0 h at different $KMnO_4$ concentrations. Low $KMnO_4$ concentrations may suffice for Cu leaching, preventing glutamate degradation, but higher concentrations can be used for faster extraction, potentially reducing capital costs in industrial applications. Similar observations were noticed by Rezaee et al., in

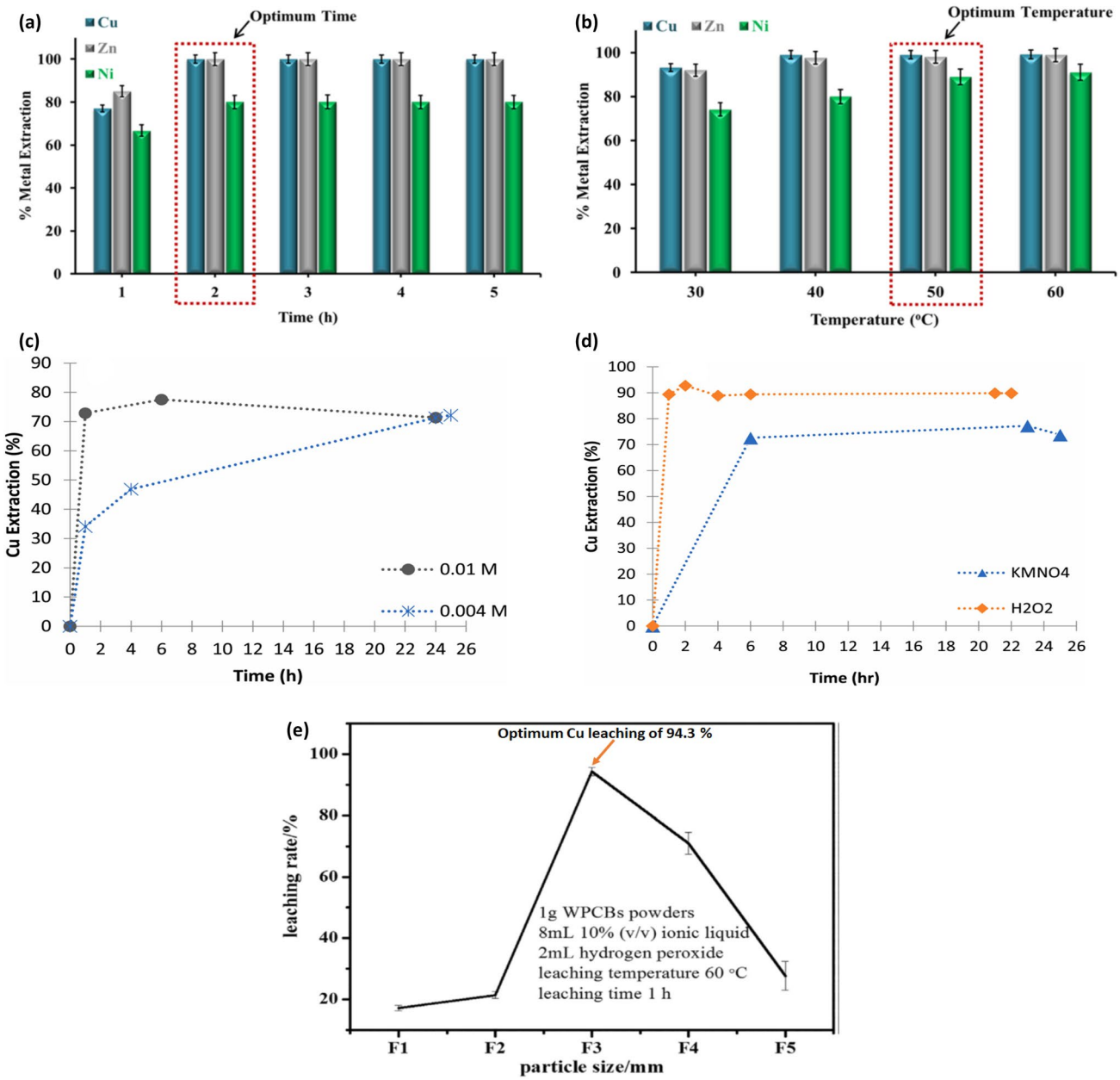


Fig. 5 Effect of various influencing parameters on the recovery of metals from DPCBs: **a** contact time, **b** temperature, reproduced from [70], Copyright 2023, with permission from Elsevier, **c** concentration of oxidants, **d** nature of oxidants, reproduced from [92], Copyright 2021, with permission from Elsevier, **e** particle size (F1 < 0.071 mm; 0.071 < F2 < 0.100 mm; 0.100 < F3 < 0.250 mm; 0.250 < F4 < 0.500 mm; and F5 > 0.500 mm), adapted from [45], Copyright 2018, with permission from Elsevier

their recent study where the most significant Au recovery occurred at a $KMnO_4$ concentration of 2.0 g/L [84]. However, when the $KMnO_4$ was increased to 4.0, 8.0, and 16.0 g/L, the final Au recovery rates decreased to 89.0, 69.0, and 32.0%, respectively.

The nature of oxidants has also been reported to affect the recovery of metals. For instance, during the recovery of Cu, as illustrated in Fig. 5(d), it was reported that the recovery platform containing H_2O_2 consistently demonstrated a higher Cu dissolution rate compared to $KMnO_4$ throughout the 24.0 h leaching study [92]. It achieved a maximum Cu extraction of 92.0% after just 2.0 h of leaching. This superior performance can be attributed to the increased reaction rate of the cathode and the presence of OH^-, which has a high standard reduction potential of 2.38 V [79, 94]. A

decline was observed in the recovery of Cu when $KMnO_4$ was employed as presented in Fig. 5(d). This decline can also be explained by the oxidation of glutamate due to the overpotential generated by the high concentration of $KmnO_4$ as mentioned earlier.

Effect of pH

The pH level plays a crucial role in the recovery of metals from PCBs regardless of the leaching agent used. Generally, the choice of pH range depends on the specific metal to be recovered. In other words, metals like Cu, Zn, Al, Au, Pb, and Ag among others exhibit better solubility and recovery rates within specific pH ranges [78, 79, 87]. For instance, an increase in the initial pH from around 8.0 to 10.0 resulted in a significant 60.0% increase in Cu extraction within 72.0 h [79]. This enhancement in Cu extraction can be attributed to the predominance of the glycinate anion at higher pH levels. Similarly, according to another study by the same research group, higher Cu and Zn extractions were achieved at elevated alkalinity (pH 11.0) [78]. In contrast, Pb exhibits better solubility at lower pH levels (pH 7.0). This finding suggests the possibility of selectively leaching Pb over Zn by adjusting the leaching pH. Furthermore, the same authors confirm that Cu, Zn, and Pb are all soluble in NH_2CH_2COOH (the leaching agent) over a wide pH range, with optimal pH levels for each metal. However, Al does not dissolve at lower pH levels due to its inability to form an aluminate complex (and it does not form stable glycinate complexes either).

Precious metals like Au and Ag, are better extracted in an alkaline medium with pH ≥ 11.0 as demonstrated by some studies [87, 95, 96]. In a 2023 study by Hao et al., Au leaching efficiency exhibited a pH-dependent behaviour [89]. Increasing the pH from 7.0 to 8.0 resulted in a significant rise in Au leaching efficiency from 76.5% to 85.9%. However, within the pH range of 8–10, Au leaching efficiency decreased notably from 85.9% to 49.7%. This pH sensitivity was attributed to changes in the stability of the Co(II)-glycine complex, a key factor in Au oxidation. Furthermore, higher pH levels promoted the dissolution of Ag and Cu, leading to increased thiosulfate consumption and a reduction in available free thiosulfate. These combined factors contributed to the decline in Au leaching efficiency at elevated pH values. Consequently, the study identified the optimal pH for Au leaching as 8.

Effect of solid/liquid ratio

The S/L ratio, which represents the amount of PCB material to the leaching solution, has a notable impact on the recovery of metals from PCBs. The choice of S/L ratio is crucial, as an excessive amount of PCB material may result in inefficient mass transfer and reduced metal recovery. Conversely, insufficient PCB material may lead to a diluted leaching solution, lowering metal concentration and recovery. In general, the leaching of metals shows a notable increase when the S/L ratio is reduced until a certain threshold is reached [19, 70, 97]. For instance, as the S/L ratio decreases from 1/2.5 to 1/15, the Cu leaching rate increases substantially from 11.9 to 87.2%, and the Zn leaching rate rises from 9.6 to 73.4% [46]. Similarly, there was a substantial increase in Cu leaching, rising significantly from 60.0% at an S/L of 1:5 to a complete 100.0% leaching efficiency at an S/L ratio of 1:15 [19]. Reducing the S/L ratio effectively increases the volume of the leaching solution, which improves contact between the DPCBs powder and the leaching solution. This enhanced contact facilitates more efficient mass transfer, thereby accelerating the leaching of metals from the DPCBs [43, 45].

Moreover, in a recent study, the S/L ratio varied from 1/10 to 1/30 and at an S/L ratio of 1/10, Cu, Zn, and Ni extraction efficiencies were 66.0, 70.0, and 60.0%, respectively [70]. Nevertheless, when the S/L ratio was changed to 1/20 improved metal extraction, achieving complete Cu and Zn extraction and approximately 80.0% Ni extraction was observed. However, a further decrease in the S/L ratio did not significantly impact Ni extraction. Consequently, the S/L ratio of 1/20 was observed to be the optimal condition for metal extraction from DPCBs. Similar studies indicated that above a certain S/L, additional changes do not enhance metal extraction [84, 97].

Effect of particle size

Smaller PCB particle sizes tend to offer a larger surface area for interaction with leaching agents, potentially leading to more efficient metal recovery. This is due to the increased accessibility of metals within the particles. Nevertheless, there exists an optimal particle size range where metal recovery is maximised [43, 45]. This range varies depending on the specific leaching process and the targeted metals. In this range, the balance between increased surface area and manageable processing is achieved. However, extremely fine particle sizes may not always result in higher metal recovery [46]. Beyond the optimal range, diminishing returns can occur as fine particles may aggregate, leading to processing

challenges and lower recovery efficiency. For example, when the particle size of PCB was increased from <0.075 mm to 0.1–0.25 mm, the Cu leaching performance increased significantly from 51.3 to 99.2% [43]. This increase can be attributed to the larger surface area per unit mass, facilitating better contact between the leaching solution and Cu in fine particles. However, when the particle size was further increased from 0.25–0.5 mm to >0.5 mm, the Cu leaching rate decreased from 73.5 to 63.5%. Similarly, the maximum Cu yield reached was 94.3% as depicted in Fig. 5 **(e)** when the particle size was within the range of 0.100–0.250 mm [45]. However, for particle sizes larger than 0.250 mm, the Cu leaching rate showed a decreasing trend. This decline occurred because reducing the particle size below a critical level could lead to increased particle–particle collision and severe attrition. Consequently, the leaching liquid struggled to permeate through the fine DPCB powder, hindering the leaching process [98].

Conclusion and future directions

The review underscores the critical importance of responsible PCB management today. As we navigate the challenges of e-waste disposal and resource scarcity, the findings presented here emphasise the need for sustainable practices and innovative solutions in the field of PCB recycling. The integration of environmentally friendly approaches, coupled with the optimisation of leaching parameters, offers a promising pathway towards reducing the environmental burden of DPCBs and harnessing their valuable metal components.

In the field of metal recovery from DPCBs, the utilisation of ILs, DESs, organic acids, and amino acids has emerged as a dynamic frontier. However, not much information is available yet on the leaching of metals from DPCBs using DESs. Nevertheless, each of these solvent systems exhibits distinct interactions with metals and PCB, emphasising their versatility in metal recovery processes. Most of these recent leaching methods were able to leach base metals (Cu, Fe, Zn, Sn, etc.), as well as precious metals (Au and Ag), successfully with leaching efficiencies reaching more than 90.0% in most cases. Hence, their effectiveness rivals that of traditional hydrometallurgical approaches using inorganic acids such as H_2SO_4, HNO_3, HCl, etc., and pyrometallurgical methods that require high-temperature operations, as well as biometallurgical methods [30, 99]. In addition, most of the studies conducted so far reported that base metals demonstrated good recovery in an acidic medium (although some authors reported otherwise), while precious metals can be easily recovered in an alkaline medium.

The recovery of metals from DPCBs is influenced by several key parameters. The choice of leaching agent, including acids, organic acids, amino acids, DESs, or ILs, is crucial, as each agent has unique interactions with metals and PCB substrates. Additionally, the concentration of the leaching agent plays a significant role, with optimal concentrations varying for different agents and metals. The presence of oxidants like H_2O_2, I_2, $KMnO_4$, $K_3[Fe(CN)_6]$ or O_2 can enhance metal recovery by promoting oxidation reactions, but careful control of oxidant type and concentration is essential. pH levels impact metal complex solubility and stability, with varying optimal pH conditions for different metals and leaching agents. Particle size affects metal recovery, with smaller sizes offering greater surface area for leaching, but excessive attrition must be avoided. The S/L ratio, temperature, and contact time also play significant roles in optimising metal recovery processes from DPCBs.

Significant further research is needed to enhance our comprehension of the molecular interactions, molecular dynamics, and interfacial electrochemistry between diverse leaching agents (both alkaline and acidic) and metals. Also, as we look towards the future, the need for solvent regeneration becomes a pivotal aspect of sustainable metal recovery. Furthermore, conducting a comprehensive life cycle assessment study is essential to assess the environmental impacts and sustainability of the developed metal recovery technologies. Future research should aim to quantify the environmental benefits and drawbacks, allowing for informed decisions and improvements in the technology. Finally, to bridge the gap between laboratory research and practical application, pilot-scale studies are crucial. Researchers should conduct pilot-scale experiments to validate the scalability and feasibility of the developed metal recovery technologies, laying the foundation for potential industrial implementation.

Acknowledgement(s) Emmanuel A. Oke acknowledges the Research Office of the University of the Witwatersrand for the Centennial Postdoctoral Fellowship granted to him between 2023 and 2025.

Authors contributions EAO: Conceptualisation, Methodology, Data curation, Writing—Original Draft, Writing—Review & Editing, Validation, Visualisation. HP: Writing—Review & Editing, Supervision, Validation, Resources.

Funding Open access funding provided by University of the Witwatersrand. This work did not receive any specific funding.

Declarations

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

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