Eng. Sci., 2023, **22**, 844

Engineered Science
DOI: https://dx.doi.org/10.30919/es8d844

A Comprehensive Review on Sustainable Metal Recovery from E-Waste Based on Physiochemical and Biotechnological Methods

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Abstract

Numerous studies have concentrated on the recovery of valuable metals from waste printed circuit boards (WPCBs) in recent years. In this review study, metal recovery based on various physiochemical and biotechnological methods is reviewed majorly classified into five techniques: (i) physical recycling techniques, (ii) thermo-chemical techniques, (iii) pyrometallurgical method, (iv) hydrometallurgical method, and (v) bio-metallurgical method. Beneficially, these techniques favor the reusing of valuable metals from e-waste. It is found that some of the techniques such as pyrometallurgy and hydrometallurgy have been used traditionally which requires high cost and also has some harmful effects on the environment. In order to obtain socio-economic and eco-friendly methods, the development of bioleaching using various microorganisms emerged in the field of biotechnology precedes the way to recover heavy metals. Based on the literature review, some of the important and experimentally proven techniques analyzed over years have been stated in this study. The significance of recovering metals from e-waste and their applications in various fields were also discussed.

Keywords: Metal ions; Waste management; E-Waste; Sustainable recovery; Physiochemical recovery; Biotechnological recovery.
Received: 10 February 2023; Revised: 07 March 2023; Accepted: 19 March 2023.
Article type: Review article.

1. Introduction

Due to rapid technological advancements in information and computing technology, global demand for electrical and electronic items is increasing steadily.[1] This leads to the increased rate of production of novel advanced appliances that replace the former technology resulting in the abundant production of antiquated products termed WEEE(Waste Electrical and Electronic Equipments) or E-waste (Electronic waste).[2] The most prevalent E-waste among the various types is from mobile devices and laptops. In addition, it is said that India produces E-waste at a rate that is typically four times more than that of the USA.[3] The majority of the E-waste contains Printed Circuit Boards or PCBs which constitute roughly 3-6% of its overall weight. This PCB carries approximately 28% of metallic components and 72% of non-metals, plastics, and ceramics. The constituents of these elements differ according to the type of product that is produced and also depend on the choice of the manufacturers to produce a complete product.[4-7] PCBs contain precious and valuable metals such as steel, bismuth, copper, silver, aluminum, barium, iron, nickel, palladium, and gold, *etc*., They also contain many hazardous materials such as antimony, arsenic, chromium, beryllium, lead, cadmium, and mercury. Additionally, they contain Polyvinyl chloride, Polychlorobiphenyl, and polymers with Brominated Flame Retardants (bromine, chlorine, phosphorus, or nitrogen). These heavy hazardous metals may release some gasses during the recovery process and may cause a great impact on human health.[8-13] Initially E-waste has been disposed of by incineration, landfills, and acid-washing techniques however it may release some toxic substances which affect the environment. Due to these risks, the scientific community has

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developed alternate ideas to recycle electronic waste. They also potentiate great opportunities in the business field and also reduce the requirement of mining. Recycling electronic waste is an important process to eliminate the need for new products. It involves the disassembly or destruction of E-waste and the data analysis shows that only 15% are admitted for recovery, in which they are subjected to different types of metal separation processes either through chemical or biological methods.[14-16] Based on the type of metal to be recovered several conventional methods such as pyrometallurgy, hydrometallurgy, and bio-metallurgy techniques are used.[17,18] Pyrometallurgical method uses heat treatment like roasting, and smelting whereas the hydrometallurgical method uses chemical leaching by using acids or alkalis. These two methods are purely based on the chemical reaction of the desired metal that is to be separated.[11] The pyrometallurgical and hydrometallurgical methods are shown to be ecologically unfeasible because of their high cost and risk process which also have difficulty in the control and the production of secondary waste. The especially pyrometallurgical method has significant energy requirements along with smelted metal and solid waste concentrates.[19] It also has disadvantages such as low selectivity, high operating expenses in terms of capital and social acceptance, vigorous thermal treatment, and negative thermal impact. It indicates that these methods have high energy costs and carbon gas emissions which prompts the search for another conventional method.[20] The other conventional method for metal recovery is bio-metallurgy or bio-hydrometallurgy which follows the process of hydrometallurgy along with the use of microorganisms. In this bio-metallurgy process, there are five different mechanisms including biotransformation, bioaccumulation, biosorption, biomineralization, and bioleaching.[21-23] Among these, bioleaching is a simple and cost-effective method with advantages such as low energy consumption, implementation of process at atmospheric pressure and ambient temperature, environmentally safe, and not requiring specialized personnel.[24] Considering all these accounts, this study has been done to state various methods and their significance in the recovery of metals. Electronic wastes are dumped into open wells of water bodies, landfills, and agricultural fields without knowing the impact on humans and the environment. Toxic substances such as cadmium and poly-brominated flame-retardant plastics are leached into the soil which may contaminate the groundwater and the plants. Later when consumed by human beings it may become a threat to their health.[25] Exposure to these substances for a long time may lead to changes in the nervous, skeletal, excretory, reproductive, and endocrine systems of the body. To conquer this problem and to safeguard the environment from hazardous electronic waste substances certain recovery and management protocols have been introduced. These strategies not only help to eliminate E-waste from the environment but also increases the retrieval of some precious metals in a successful manner which were currently used in many fields.[26] The existence of PCBs in electronics increases the potential for recovering precious metals, for example, silver, gold, nickel, palladium, copper, platinum, and iron which captivates commercial interest. Its superior level of purity compared to metals derived from ores is primarily what makes it significant. It is also said that recycling a million cell phones can provide roughly 34kg of gold.[3] PCBs of waste computers generate about 22.7% oil, and 4.70% gasses having copper-rich residue and a high calorific value of 70% which can be used for fuel and as a substitute for energy production and also for performing some chemical reactions.[27,28] Metal recovery from electronic waste should be done through recycling to save the environment and increase the utility of used printed circuit boards.

2. Strategies for recovering metal from used printed circuit boards

The small and medium industries have gained high profits in recovering metals from e-waste. However, numerous traditional thermo-chemical and biochemical methods for metal recovery from E-waste have been tested in pilot and laboratory studies. Numerous scientists have employed various technologies to retrieve metals from electronic waste, including physical recycling techniques, thermo-chemical techniques, pyrometallurgical methods, hydrometallurgical methods, and bio-metallurgical methods. The general schematic flow of these methods is shown in Fig. 1.

2.1 Retrieval of metals by physical recycling process

Physical recycling methods are the preliminary method in metal recovery from electronic waste. They generally liberate numerous metals and non-metals in an efficient manner so it is used as an appreciable technique for recycling electronic waste globally. Before further processing, it includes steps like dismantling, disassembling, shredding, chopping, crushing, *etc*. This approach is also regarded as a pre-treatment phase. These procedures are carried out using equipment such as granulators, pre-granulators, shredders, *etc*. The separation of metals from non-metals can be achieved by using a variety of techniques, including milling, froth flotation, corona discharge, eddy current separation, density separation, and magnetic separation.[29-31]

2.2 Retrieval of metals by thermo-chemical methods

Pyrolysis is the major thermo-chemical process in which there is thermal degradation in the absence of air on the targeted E-

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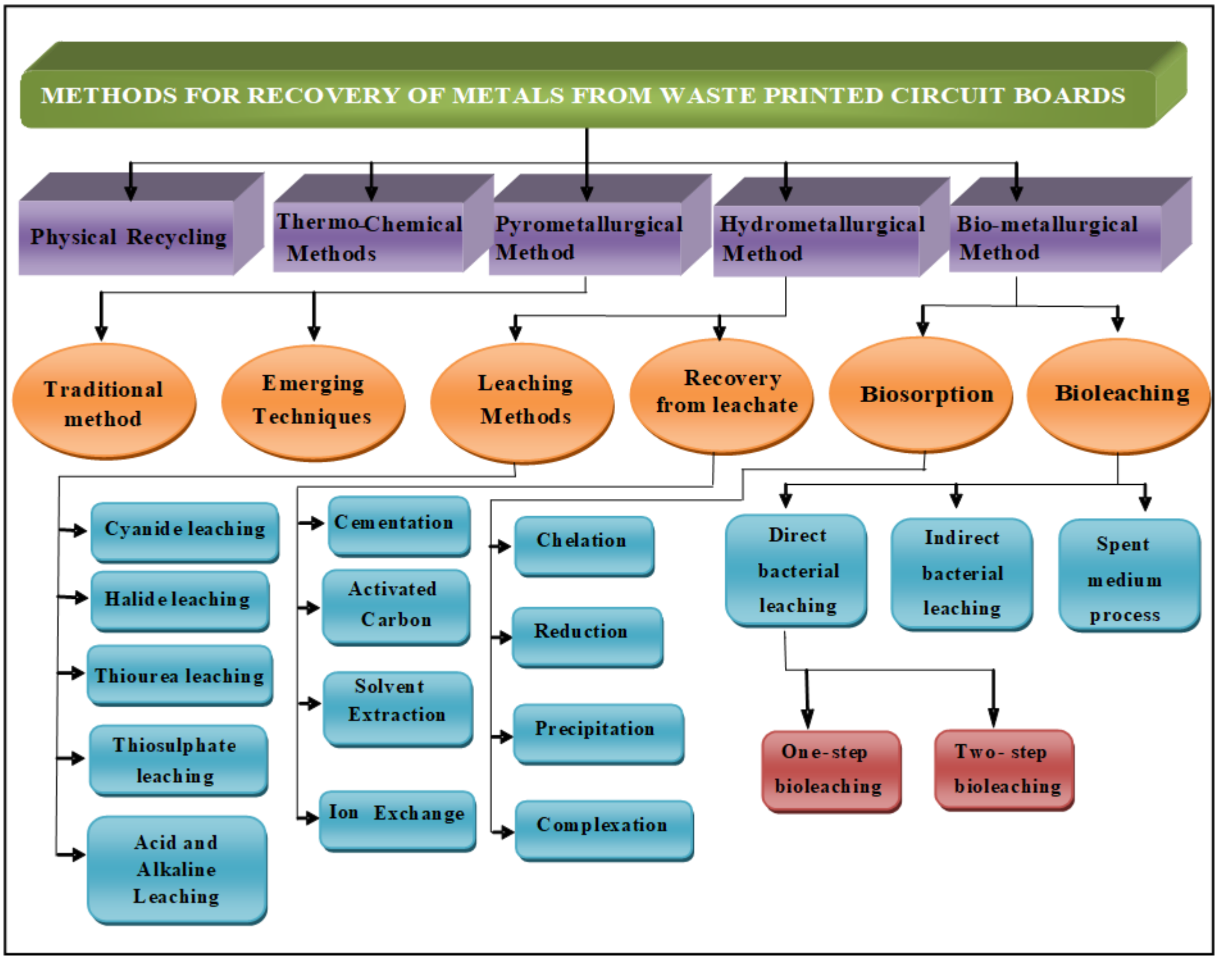

Fig. 1 Methods for retrieving metals from WPCBs.

waste material. Numerous investigations have been accomplished on the pyrolysis technique. It involves vacuum pyrolysis, microwave-induced pyrolysis, catalytic pyrolysis, and co-pyrolysis. However, pyrolysis has been implemented in treating waste and recovering metals, novel plasma technological method has gained attention over the other processes. This plasma technology method is applicable to biomedical waste disposal.[30,32]

2.3 Retrieval of metals by the pyrometallurgical method

The pyrometallurgical method is the widely employed process due to the easy recovery of metals from complex PCBs. The PCBs are shredded into fine pieces before pyrometallurgical processing. In this method smelting, refining, incineration, and combustion is the familiar process.[33] High temperatures in melting furnaces combined with unique melting fondants increase the effectiveness of this approach. A temperature of about 400 to 700 °C was applied to the sample. After some time, materials like rubber, paper, wood, and plastics break down and release volatile compounds that can be employed in the chemical industry. They can also be collected in the form of gases or oils that can be used to produce energy and carry out other chemical reactions.[34] Smelting is preceded by electrochemical refinement in this pyrometallurgical procedure. Before being fed into the furnace, where metals are recovered in a molten bath and oxides are removed from the slag phase, e-waste is physically recycled. Aurubis Recycling, OutotecTSL, and Umicore are a few companies that use pyrometallurgical processes to recover metals from e-waste.[33]

2.3.1 Pyrometallurgical Technique for metal recovery from E-waste

Boliden Ltd. Rönnskår Smelter in Sweden employed a pyrometallurgical process to retrieve metals from electronic scraps. The scrap E-waste is fed into the process in stages based on its purity. Scarp containing a high copper concentration is fed directly into the conversion process, whereas low-grade E-waste is fed into the Kaldo furnace. A skip hoist powers the Kaldo furnace, and an oil-oxygen burner provides the necessary amount of oxygen for burning. Off gases are also subjected to post-combustion in the furnace at 1200 degrees Celsius. As a result, the Kaldo furnace produces a heterogeneous copper mixture including silver, copper, gold, zinc, nickel, palladium, selenium, lead, cadmium, indium, and antimony dust.[35] The schematic representation of the whole process in Rönnskår Smelter is given in Fig. 2.

Umicore developed a novel metal refining process in Hoboken, Belgium, aimed at recovering the majority of valuable metals from e-waste. Metal sweeps and bullions, wasted industrial catalysts, and printed circuit boards were all

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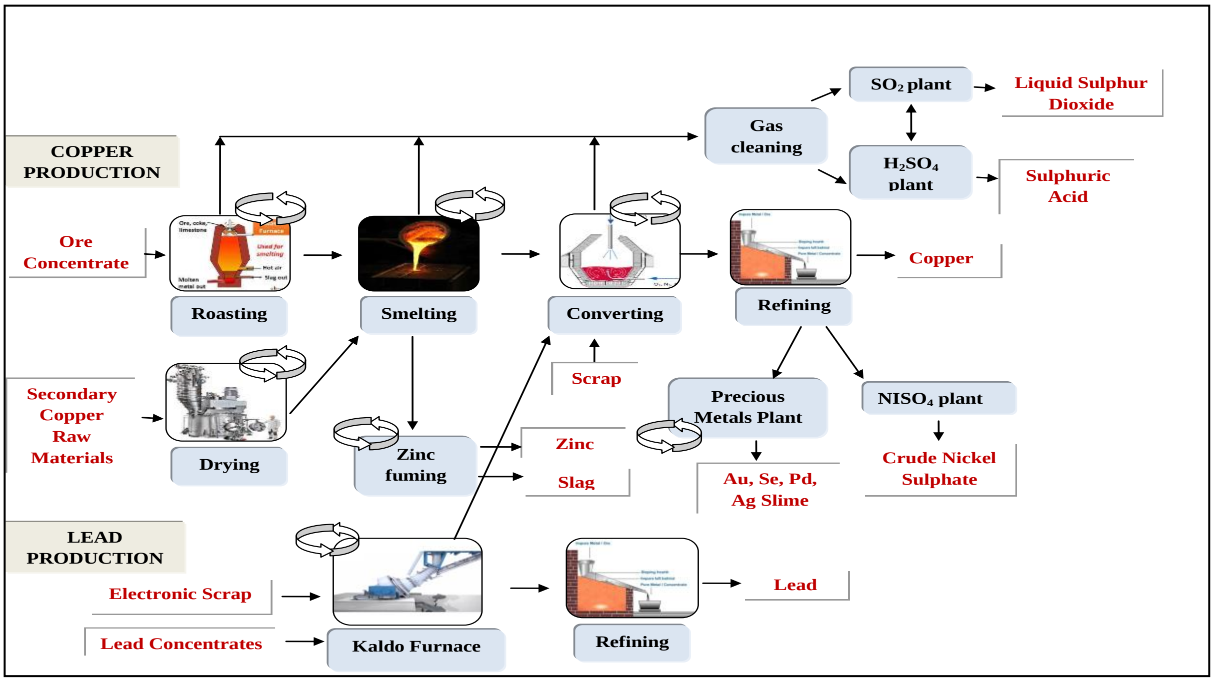

Fig. 2 Schematic representation of Rönnskår Smelter Process.

appropriate for the metal smelter and refinery processes. The first phase is carried out using the Isa Smelt furnace. Other organic chemicals and polymers in the feed operate as a reducing agent and a source of energy in part. All other metals are concentrated in a lead slag by the smelter, which separates them into copper bullion before processing them at the base metals operations (BMO). To recover precious metals from copper bullion, copper-leaching, and electro-winning are used.[36] A diagrammatic representation of Isa-Smelt furnace is shown in below Fig. 3.

2.3.2 Emerging advanced technique in pyrometallurgical process for recovery of metals

Metal recovery by pyrometallurgical methods is becoming more common. Boliden Minerals AB, the Association of Plastics Manufacturers Europe (APME), and the American Plastics Council (APC) undertook research that introduced the recovery of zinc from slag via the zinc fuming method. The PCB remnants were collected and the Hg-containing portions were dismantled and crushed into fine fragments. The crushing mill and ferromagnetic separator were used for pre-treatment. Shredded scrap with particle sizes of 3cm was combined 50:50 with shredded reversion material to maximize bulk processing and silo feeding while preventing clog throughout the input. Rich metals and copper can be recovered from e-waste by a copper smelter. This procedure is said to allow the complete recovery of Cu and other metals.[35]

Dunn *et al*. discovered how to recover precious metal gold from gold scrap. At a temperature of around 300-700 °C, gold scrap interacts with chlorine to generate a metallic complex comprised of gold, chloride, silver, and various metallic chlorides. The final combination was treated using air-sparged HCl to dissolve the poor metallic chlorides besides silver chloride, resulting in a silver chloride / metallic gold mixture. The silver chloride / metallic gold mixture was then dissolved with NH_4OH and HNO_3 to get 99.9% pure gold retrieved from gold trash.[37] Day proposed a novel method for retrieving valuable metals from porous ceramic substances. The waste was fed into a plasma arc furnace at 1400 °C, producing two metallic phases: a molten phase containing a mixture of valuable metals and collector metal, and a slag phase containing ceramic remnants. If required, divided silver or copper are excellent collector metals. This process recovers 80.3% of platinum and 94.2% of palladium, respectively.[38] The above-mentioned pyrometallurgical metal recovery methods and their features are indexed in Table 1.

2.4 Retrieval of metals by the hydrometallurgical method

When compared to other pyrometallurgical procedures, hydrometallurgy is the chemical leaching process that is more predictable and controllable. To dissolve waste for the pre-treatment process uses acids or ammoniacal solutions such as aqua regia, sulphuric acid (H_2SO_4), hydrochloric acid (HCl), and a few alkalis.[48] The impurities have been removed using the gauge materials and the metals are isolated by adsorption, solvent extraction, ion exchange, *etc*. The final form of metal is retrieved either by the electro-refining or chemical reduction process.[33,49]

2.4.1 Recovery by leaching methods

Leaching is the major hydrometallurgical method in which the soluble constituent is separated by using solvent as an initial process from the solid component. The most widely used leaching agents are cyanide, thiourea, halide, thiosulfate, acid, and alkaline compounds.[50-53]

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Table 1. Tabulation on types of pyrometallurgical methods.

Pyrometallurgical Techniques	Recovered Metals	Features of Main Process	Results	Reference
Boliden Rönnskår Smelter	Ag, Se, Au, Zn, Pd, Cu, Pb, Ni	Lead concentrates are fed into the Kaldo reactor.	Copper and other precious metal recover at a high rate	[39, 40]
Noranda process	Ag, Pt, Pd, Se, Au, Te, Cu, Ni	The copper converter and refining process has been upgraded; feeding copper concentrate (14% concentration) to copper smelter; recovering metals by electro-refining	Copper and other valuable metals both rebound well.	[41]
Umicore's refining process of metal	Precious and base metals, Indium, Tellurium, Selenium, and platinum group metals	Excellent recommendation Precious Metals Operation (PMO) includes Isa Smelt, electro-winning, copper leaching, and precious metal refinery; Plastics are used as replacements, and coke is used as a reducing agent. Isa Smelt incorporates Offgas emission control. There is also a lead refinery and furnace, and a special metals unit.	Retrieval of valuable metals such as Te, Bi, Sb, Sn, In, and Se.	[42, 43]
Umicore smelter's full-scale trail	Metals present in e-waste	WEEE-derived plastics were studied as a source of energy for the IsaSmelt to replace Coke.	The substitution of 6 percent WEEE polymers and 1 percent coke for 4.5 percent coke has no detrimental effects on smelter operation or metal recovery.	[44]
Day's patent for precious metal refractory ceramic fragments	Palladium and Platinum	The electronic scrap was charged to 1400 ºC temperature at the plasma arc furnace.	Platinum and Palladium recovered at80.3% and 94.2%	[45]
Dunn's gold refining process	Au	Gold scrap is treated with chlorine at temperatures ranging from 300°C to 700°C; HCl is used to dissolve impure chlorides. Washing withNH_4OH and $HNO_3$to disintegrate the AgCl	99.9% pure gold recovered from scraps.	[46]
Aleksandrovich's Patent for PGM and Au Recovery from e-waste	PGM and Au	Fuse wastes depending on base metal chalcogenides with a carbon reducer; Melted materials are cooled, solidified, and solidified product separation takes place through created phase boundaries.	PGM and gold retrieved from scraps.	[47]

a) Cyanide leaching

In the mining sector, cyanide functions as a gold-leaching agent. The electrochemical process explains the dissolving mechanism of gold from cyanide solution, which comprises the following processes as expressed in equations (1) and (2).

$$4Au_{(s)} + 8CN^{-}_{(l)} \rightarrow 4Au(CN)_{2(l)}^{-} + 4e^{-} \quad (1)$$

$$O_{2(g)} + 2H_2O_{(l)} + 4e^{-} \rightarrow H_2O_{2(l)} + 4OH^{-} \quad (2)$$

A diagrammatic representation of the mechanism of cyanide leaching is shown in Fig. 4.

This reaction has been affected by pH due to the dissolution rate of the different types of metals. In a study, it is proved that in a cyanide solution, maximum dissolution of silver, gold, platinum, and palladium might be achieved at the pH 10-10.5 which follows the activity in the order of gold > silver > palladium > platinum. However, the use of cyanide as a leaching agent may cause severe contamination to the ground water becomes the major reason for the non-cyanide substitutes such as thiosulfate and thiourea.[54,55]

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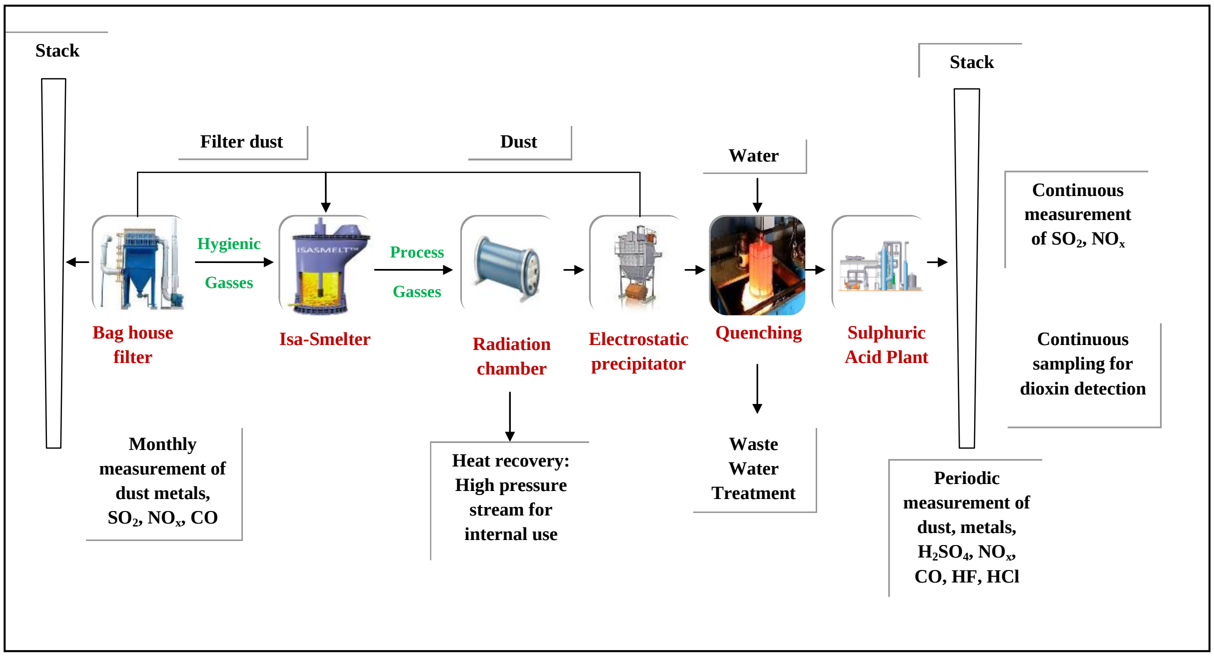

Fig. 3 Diagrammatic representation of Offgas emission control in Isa-Smelt furnace.

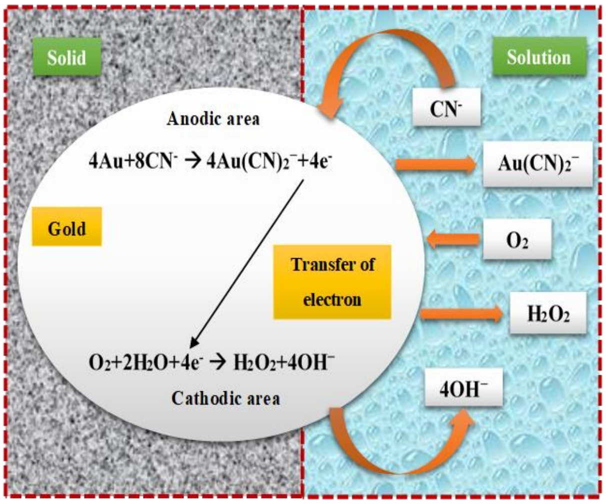

Fig. 4 Diagrammatic representation of cyanide leaching.

b) Halide leaching

Chlorine, iodine, bromine, fluorine, and astatine are halides that are employed as a lixiviant for dissolving metals, particularly gold. Except for fluorine and astatine, all of the halogens described above have been subjected to gold recovery, where they form Au(I) and Au(III) complexes with iodide, bromine, and chloride. Among these halogens, chloride is largely applied on the industrial scale. The chlorination rate has been increased by low pH, high chlorine, or chloride content with increased temperature and surface area. Aqua regia, a combination of 3 parts of Conc. HCl and 1 part of Conc. HNO_3 has been traditionally used as a dissolving agent for gold. The overall reactions are as follows represented in equations (3) and (4):

$$2HNO_{3(l)} + 6HCl_{(l)} \rightarrow 2NO_{(g)} + 4H_2O_{(l)} + 3Cl_{2(g)} \quad (3)$$

$$2Au_{(s)} + 9HCl_{(l)} + 3HNO_{3(l)} \rightarrow 2HAuCl_{4(l)} + 3NOCl_{(l)} + 6H_2O_{(l)} \quad (4)$$

Chloride leaching can be hard to implement for gold extraction than cyanide leaching for two reasons.

• Special stainless steel and rubber-lined equipment are required to withstand the highly corrosive acidic and oxidizing conditions.

• Since chlorine gas is extremely dangerous, it should be monitored to avoid health risks.[15,52,56]

c) Thiourea leaching

Thiourea ($(NH_2)_2CS$) is used as a gold extraction agent from ores. Thiourea dissolves gold in an acidic environment and creates a cationic complex, resulting in gold extraction of up to 99%.[54] The overall reaction to thiourea leaching is as follows in equation (5):

$$Au_{(s)} + 2CS(NH_2)_{2(l)} \rightarrow Au(CS(NH_2)_2)^{2+}_{(l)} + e \quad (5)$$

This process is primarily governed by

(a) Thiourea and electron recipient concentrations, which enhance the rate of leaching

(b) Utilization of Fe^{3+} in H_2SO_4

(c) Rate of dissolution of gold is heavily influenced by pH.

This method has proved the effectiveness of thiourea, however, this process has been hindered due to the following reasons includes

(i) Most expensive than the cyanide method

(ii) The consumption of thiourea is high due to its fastest oxidization in gold processing

(iii) Thiourea procedure is still in its infancy, and the recovery approach requires additional steps.[57]

d) Thiosulfate leaching

Many researchers employ thiosulfate ($S_2O_3^{2-}$) as a leaching agent in the pharmaceutical and photography industries as asbustitute for cyanide. The presence of cupric ions causes gold disintegration locally. The $Cu(NH_3)_4^{2+}$ions in solution

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obtain electrons from the cathodic portion of the gold, which is subsequently reduced to $Cu(NH_3)_2^{+}$. Au^{+} ions on the anodic surface combine with ammonia or thiosulfate ions as they enter the solution to form $Au(NH_3)_2^{+}$ or $Au(S_2O_3)_2^{3-}$, respectively. Depending on the concentration of $S_2O_3^{2-}$, $Cu(NH_3)_2^{+}$ transforms to $Cu(S_2O_3)_3^{5-}$ ions, and $Au(NH_3)_2^{+}$ accomplishes the same. $Cu(S_2O_3)_3^{5-}$ and $Cu(NH3)^{2+}$ in the solution are then oxidized with oxygen to form Cu(NH3)42+. The role of copper(II) ions in the oxidation of metallic gold to Au+ ions is summarized in equations (6) and (7):

$$Au_{(s)} + 5S_2O_3^{2-}{}_{(l)} + Cu(NH_3)_4^{2+}{}_{(l)} \rightarrow Au(S_2O_3)_2^{3-}{}_{(l)} + 4NH_{3(l)} + Cu(S_2O_3)_3^{5-}{}_{(l)} \quad (6)$$

$$2Cu(S_2O_3)_3^{5-}{}_{(l)} + 8NH_{3(l)} + 1/2O_{2(g)} + H_2O_{(l)} \rightarrow 2Cu(NH_3)_4^{2+}{}_{(l)} + 2OH^{-}{}_{(l)} + 6\,S_2O_3^{2-}{}_{(l)} \quad (7)$$

The diagrammatic representation of the reaction mechanism of thiosulfate leaching is shown in Fig. 5.

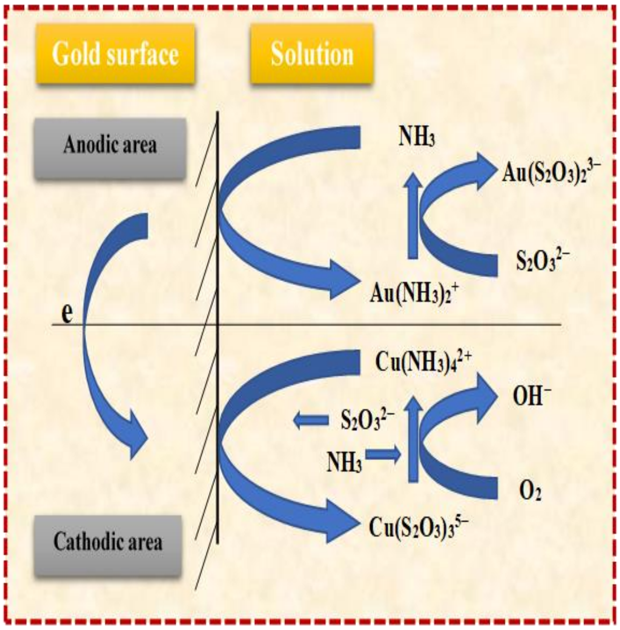

Fig. 5 Diagrammatic representation of thiosulfate leaching.

This method produces a less stable gold thiosulfate complex and requires an alkaline condition to prevent the decomposition by acid. This complex requires a pH of 9-10 and a temperature of about 25 °C otherwise it will get decomposed naturally.[15] Thus the various methods of the hydrometallurgical process and their conditions, advantages, and disadvantages are mentioned below in Table 2.[58]

e) Acid and Alkaline Leaching

Nitric acid, sulphuric acid, and hydrochloric acid are the three main mineral acids now being studied for their potential application in recovering the targeted metals.[6,59,60] The removal of some light metals, particularly from worn batteries and electronic equipment, involves the use of a few organic acids, such as citric acid, ascorbic acid, and occasionally acetic acid. Printed circuit boards typically have an inert chemical layer that prevents lixiviants from interfering chemically. Applying sodium hydroxide initially removes that layer.[61] This process eliminates cobalt and lithium, which are commonly found in lithium-ion batteries (LIBs). Moreover, the electrode plates of LIBs may have nickel and aluminum traces.[62,63] 90% of cobalt was recovered after treatment with citric acid and hydrogen peroxide (H_2O_2) as reducing agents from used LIBs, which also led to the complete recovery of Li. Hydrochloric acid was used as the leaching agent and hydrogen peroxide as a reducing agent in similar research to leach off full Li from old batteries. Leaching rates were unaffected by changes in reducing agent concentration, although it was shown in that investigation that when temperature and hydrochloric acid concentration increased, leaching efficiency improved initially before declining. The leaching efficiency was also improved by extending the contact time and prolonging the leaching period, respectively.[62,64] In order to maximize the effectiveness of organic acid leaching from e-waste, H_2O_2 is frequently employed in combination with acids.

2.4.2 Recovery of Metals from Leachate

Metals can be recovered from these varied solutions utilizing various techniques such as ion exchange, solvent extraction, cementation, and activated carbon adsorption.

Table 2. Comparison of leaching methods in the Hydrometallurgical process.

Lixiviants	Reagents	Conditions	Advantages	Disadvantages
Cyanide	CN^{-}, O_2	pH> 10 25 °C	An effective method for gold recovery	High toxicity
Thiosulfate	$S_2O_3^{2-}$, NH_3, Cu^{2+}	pH> 8-11 25 °C	The inexpensive, eco-friendly, rapid leaching rate	Downstream metal recovery, is less stable
Thiourea	$CS(NH_2)_2$, Fe^{3+}	pH = 1-2 25 °C	Fast leaching rate	Low chemical stability, a potential carcinogen
Halide	Br/Br_2, Cl^{-}/Cl_2, I^{-}/I_2	pH<4 25 °C	Relatively high stability, safety, and low cost	Hard to implement, early stage of development
Acid	H_2SO_4, HNO_3, HCl, citric acid, acetic acid	pH<3 25 °C	High and fastest metal recovery	Not simple and can produce toxic products
Alkaline	NaOH	pH>8 25 °C	Applied to large operations	Extraction of metals is low

a. Metal recovery by Ion-Exchange

Zhang and Dreisinger used ion exchange columns to recover gold from ammoniacal thiosulfate solutions containing copper. Amberlite IRA-410, Dowex 21K, and Dowexwere was chosen from the group of resins. These gel resins contain quaternary ammonium functional groups and a polystyrene divinylbenzene matrix. According to the findings, gold can be fed onto strong basic ion exchange resin from thiosulfate solutions in the absence of copper to high loading concentrations, whereas in the presence of copper, ion exchange operation is only effective under limited conditions due to less stability and the possibility of forming poisonous polythionates.[65] Lewatit TP 207 resin selectively separated Ni (II) from spent HDS catalyst ammonia leaching solution in another ion exchange study. This process may be considered effective for the recovery of metals and relevant to the industry due to the reduction in the discharge of liquid effluent or solid waste. Ion exchange resins also play an indirect role in metal recovery from discarded catalysts.[66, 67]

b. Metal recovery by solvent extraction

To address the demands of gold extraction, metal recovery by solvent extraction has been investigated. Numerous studies were carried out on extraction systems including organophosphorus derivatives, a mixture of amines-organophosphorus, and guanidine derivations derivatives. From the thiosulfate solution, the extraction of gold was done with the help of alkyl phosphorus esters. NH_3 has a significant impact on the recovery of Au from thiosulfate solutions containing alkyl phosphorus esters. The reaction was expressed as follows in the equation (8) and (9) for the system with no ammonia:

$$i\mathrm{Na}_{(l)}^{+} + 2\mathrm{Au}^{+}_{(l)} + j\mathrm{S_2O_3}^{2-}{}_{(l)} + \mathrm{OH}^{-}_{(l)} + m\mathrm{TBP(o)} = \mathrm{Na}_i\mathrm{Au_2(S_2O_3}^{2-})_j(\mathrm{OH}^-).m\mathrm{TBP(o)}_{(l)} \quad (8)$$

When ammonia was added to thiosulfate solutions, the reaction was represented as:

$$i\mathrm{Na}^{+}_{(l)} + 2\mathrm{Au}^{+}_{(l)} + j\mathrm{S_2O_3}^{2-}{}_{(l)} + \mathrm{OH}^{-}_{(l)} + 2\mathrm{NH_3} + m\mathrm{TBP(o)} = \mathrm{Na}_i\mathrm{Au_2(S_2O_3}^{2-})_j(\mathrm{OH}^-)(\mathrm{NH_3})_2.m\mathrm{TBP(o)}_{(l)} \quad (9)$$

where I (I = 3–5) changes with j(j = 2–3), and the TBP coefficients m in the two equations are 1.5–2.5 and 6–9 respectively.[68]

Many studies have been performed for solvent extraction recovery and separation of precious metals from spent catalysts. For example, Mo(VI) and Co(II) were extracted from the leach chloride solution of spent hydrodesulfurization (HDS) catalysts using Trioctylphosphine oxide (TOPO) and Triisooctylamine. The advantage of this technique is that theorganic phase in solvent extraction can be regenerated numerous times, adding economic value to the solvent extraction process.[67]

c. Metal recovery by cementation

Zinc cementation is done for the recovery of gold worldwide, since 1890. This process is also called as Merill-Crowe process.[69] Cathodic deposition of gold and anodic degradation of zinc on the surface of zinc particles are used to recover gold.

$$2\mathrm{Au(CN)_2}^-{}_{(l)} + 2e \rightarrow 2\mathrm{Au}_{(s)} + 4\mathrm{CN}^-{}_{(l)} \quad (10)$$

$$\mathrm{Zn}_{(s)} + 4\mathrm{CN}^-{}_{(l)} \rightarrow \mathrm{Zn(CN)_4}^{2-}{}_{(l)} + 2e \quad (11)$$

If the cyanide concentration is reduced excessively, zinc particles may develop a layer of zinc hydroxide on their surface, as described in equations (10) and (11). It stated that gold recovery is possible at pH levels ranging from 8 to 11. Nickel, copper, lead, sulfur, antimony, and arsenic are all impure metals that are unsuitable for cementation. Gold is recovered from thiocyanate, thiosulfate, and thiourea solutions via reduction precipitation with a stabilized form of sodium borohydride (SBH). This SBH is a 12% sodium borohydride and 40% sodium hydroxide aqueous solution. Pure metallic Au can be obtained by the addition of SBH to the acidic solutions even at ambient temperature. Thiourea is employed as a leaching as well as a gold stripping agent in loaded solvents and resins. The conclusion is that gold can be recovered from the acidic thiourea solution by varying the amount of reagent used.[70]

d. Metal recovery by activated carbon

McQuiston and Chapman invented this process in 1946 for the recovery of gold and silver cyanide complexes on activated carbon. New advancements in this technique have resulted in the development of the carbon-in-pulp (CIP) and carbon-in-leach (CIL) processes for the recovery of gold from cyanide solutions.[71,72] Many researchers have investigated the mechanism and kinetic adsorption of gold onto activated carbon. For solid/liquid adsorption processes, two isotherms were well- known as shown in the equation (12) to (15):

One is Langmuir isotherm,

$$Q = \frac{QmaxbC}{(1 + bC)} \quad (12)$$

where Q is the amount of solute adsorbed per unit weight of carbon, Qmax is the maximal sorption capacity, C is the equilibrium concentration of adsorbate in solution, and b is the adsorption equilibrium constant. In a multicomponent system (i=1, 2,..., k), the Langmuir isotherm equation is denoted as:

$$Q_i = \frac{bi0Ci}{(1+\sum_{j=1}^{k} \mathrm{bjCj})} \quad (13)$$

in which Qi denotes the quality of solute i, adsorbed per unit weight of carbon at equilibrium concentration, Ci in a solution containing k solutes, bi0 = Qmaxi·bi, bi is the adsorption equilibrium constant for solute i.

The Freundlich isotherm is expressed as,

$$Q = KC^n \quad (14)$$

K and n (0 <n < 1) are the Freundlich constants. In this context, a linear relationship between ln$Q$ and ln$C$ is expected. Whereas Sheindorf *et al*. established the Freundlich-type multicomponent isotherm as:

$$Qi = k_i C_i \left(\sum_{j=1}^{k} a_{ij} c_j\right) n_i - 1 \quad (15)$$

where $a_{ij}$is the competition coefficient (a_{ij} = 1). The multi-component system is used to calculate K_i = $q$miAi and K_i.[73-75]

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2.4.3 Commonly used technique for metal recovery from E-waste in hydrometallurgy

The most frequent recycling process for electronic scrap in hydrometallurgy was blast furnace smelting with secondary copper and lead smelters. Quinet *et al*. extracted valuable metals from discarded smartphones on a bench scale. The base metal is composed of 0.52% silver, 0.04% palladium, 27.37% copper, and 0.06% gold. This waste material was shredded or crushed using a Retsch heavy-duty cutting mill, and the crushed matter was divided into four particle size groups: +1.168 mm, 1.168 + 0.6 mm, 0.6 + 0.3 mm, and 0.3 mm. The shortest fraction was then investigated using a number of hydrometallurgical procedures, including cementation, precipitation, ion exchange, chloride leaching, sulfuric acid leaching, thiourea leaching, and adsorption with activated carbon, which results in a recovery of approximately 99% of the palladium, 95% of the gold and93% of the silver.[50] The schematic flow of the above-mentioned process is shown in Fig. 6.

Amino acids are currently being utilized to dissolve base and precious metals in alkaline environments, resulting in high yields. In Australia, glycine has been recently investigated as a greener alternative to traditional leaching reagents for extracting base metals from PCBs. By combining glycine and cyanide, a significant amount of copper (99.1%), gold (92.1%), and silver (85.3%) were extracted. Amino acids, including glycine, are advantageous due to their non-toxic, non-volatile, and low-cost properties, they can be produced at an industrial scale through chemical or biological synthesis.[76]

2.5 Retrieval of metals by a bio-metallurgical method

Recovering E-waste using a bio-metallurgical method provides numerous potentials for research and development as well as a lucrative economic field. This process is divided into 2 parts: a) Biosorption and b) Bioleaching. This method is solely reliant on metal adsorption from adsorbents. Biosorption of e-waste is done by using fungus (*Aspergillus niger*), algae (*Chlorella vulgaris*) bacteria (*Peniciliumchrysogenum*), alfalfa, ovalbumin, and hen eggshell membrane. The following factors influence this process: a) Biosorbent binding type (living, non-living), b) Categories of biological ligands available for metal, c) Chemical, stereochemical, and co-ordination properties of the targeted metals, and d) Metal solution characteristics such as pH and competing ions.[77] Bioleaching is the biological oxidation process in which autotrophic bacteria, heterotrophic bacteria, and fungi are used. It involves the following process such as (i) acidolysis, the solubilization of the material as a result of the acidity; (ii) complexolysis, which involves the complexation of metals with excreted organic acids or amino acids; (iii) redoxolysis, in which oxalic acid is a mediator in the reduction of ferric iron; and (iv) bioaccumulation, in which the mycelium serves as a sink for the metal ions.[78]

2.5.1 Biosorption

The biosorption mechanism is a complicated process that involves the adsorption of sorbate to the biosorbent. Numerous natural substances can operate as biosorbents by binding metal ions through physical (Vander Waals forces or electrostatic interaction), chemical (displacement of bound metal cations (ion exchange) or protons), or biological (chelation, reduction, precipitation, and complexation) processes. Chemical and functional groups in biosorbents, such as amide, amine, imidazole, sulfonate, carbonyl, thioether, sulfhydryl, phosphodiester, phenolic, carboxyl, and imine groups, can draw and bind metal ions.[79,80] This method has several

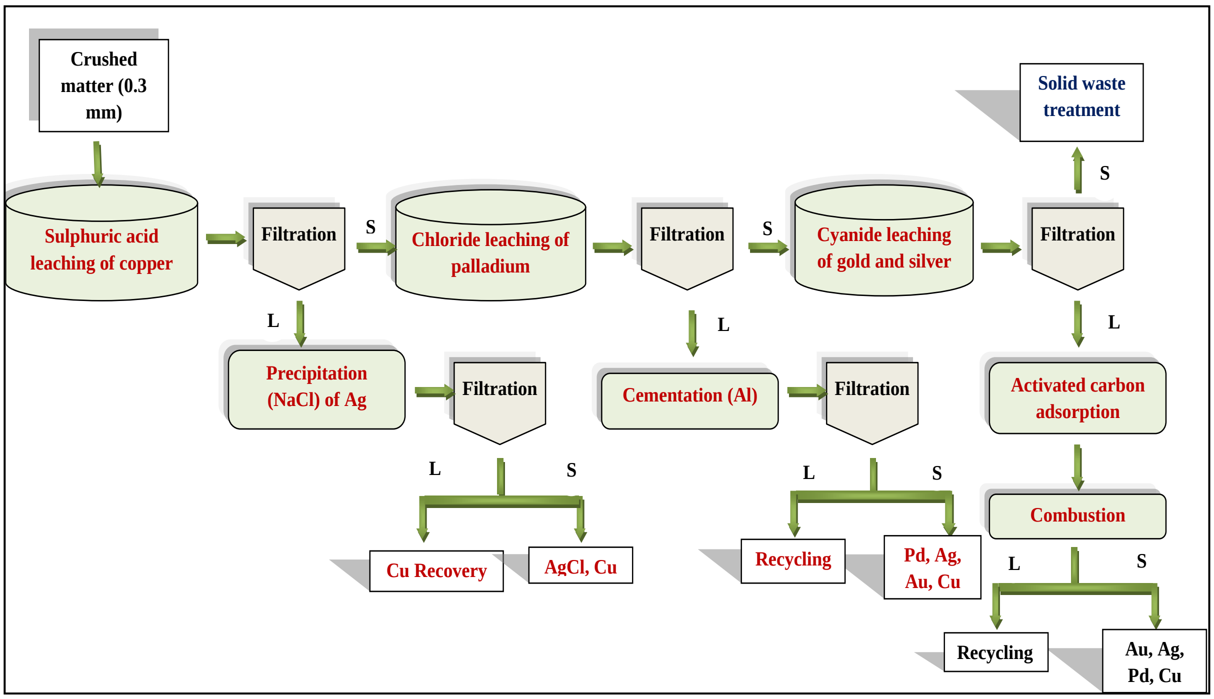

Fig. 6 Flowchart for precious metal recovery from electronic waste.

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advantages such as low cost for the production of biomass, no need for the addition of chemicals, and functions over a wide range of pH, temperature, and metal ions. The main drawbacks, however, included reversible metal sorption on biomass and saturation of active sites of metal binding ligands.

a. Chelation

Chelate refers to the complex formed when a chelating chemical attaches to a metal ion simultaneously in several sites, forming a ring structure. Because the metal ion can attach to chelates in several locations, they are more stable than complexes. For the purpose of removing Cd (II) from the effluent, rice straw was employed as a potential biosorbent. The functional groups C=C, O-H, and C-O as well as the carboxylic acids that are present on the surface of the biosorbent chelate with the biosorbed Cd (II). The removal of Cr (III) and Cu (II) by carboxyl and hydroxyl groups from the effluent also utilizes a similar process of biosorption.[81]

b. Reduction

Through interactions with functional groups like carboxyl, the metal undergoes a reduction in this process, which promotes crystal formation. By using the reduction process, metals like gold and palladium have been obtained. Once the metal attaches to the biosorbent at specific locations, it is reduced. By using the reduction method, harmful hexavalent chromium can be eliminated. Numerous species reduce Cr (VI) in the aqueous solution to Cr (III) by biosorption.[82]

c. Precipitation

Metal ions produce precipitates on the surface of microbial cells, which either stay outside or enter the microbial cell. In most cases, insoluble inorganic metal precipitates form. The use of microbial cells may lead to the formation of organic metal precipitates. The majority of the extracellular polymeric compounds excreted by bacteria contribute to the formation of organic precipitates. Cu (II) precipitation on *Mesorhizobium amorphae* results in cell surface deformation, aggregation, and destruction, according to SEM-EDX analysis.[83,84]

d. Complexation

It is described as the emergence of a complex as a result of the coexistence of two or more species. The metal ion and the ligands interact to form mononuclear complexes with the metal atom at their center. The metal atom at the center of a polynuclear complex may have a positive, negative, or neutral charge depending on the number of binding ligands present. After the biosorption of antimony (III), *Cyanobacterium microcystins* underwent attenuated total reflection infrared spectroscopy (ATR-IR) investigation, which showed the participation of hydroxyl, carboxyl, and amine groups through surface complexation.[85] Other research using *Termitomycesclypeatus*, *Acidiphilium*, and alkali-modified sewage sludge for Cd removal revealed a similar mechanism of biosorption.[81]

2.5.2 Bioleaching

Bioleaching is the microbial leaching process that uses the metabolic activity or the products of the microbes at low temperatures and mild pH. Bioleaching employs a variety of microorganisms, the most prevalent of which being sulfur and iron-oxidizing chemolithotrophic bacteria(*A. thiooxidans*, *Ferrobacillus, Leptospirillum ferrooxidans, A. ferrooxidans*), moderately thermophilic bacteria(*Sulfobacilllus thermosulfidooxidans, Thermoplasma acidophilum, S. thermosulfidooxidans*), cyanogenic bacteria(*Chromobacteriumviolaceum*), microscopic fungi (*Penicillium simplicissimum, Aspergillus niger, A. Ficuum)* and heterotrophic bacteria (*Bacillus, Pseudomonas,* S*ulfolobus*).[86]Bioleaching is a generally simple procedure to operate and environmentally sustainable than other traditional extraction methods. It can able to extract metals that are too poor to extract from the other conventional methods. Over these advantages there are certain disadvantages is that it is time-consuming compared to other methods. However, bioleaching can be classified into the following types:

a. Direct bacterial leaching

It is the process by which a microbe binds to the waste's base and oxidizes the element contained in the E-waste through a series of enzymatic catalysis processes.[87] The process is given in equations (16) and (17):

$$4FeS_{2(l)}+14O_{2(g)}+4H_2O_{(l)}\rightarrow bacteria\rightarrow 4FeSO_{4(l)}+4H_2SO_{4(l)} \quad (16)$$

$$4FeSO_{4(l)}+O_2+2H_2SO_{4(l)}\rightarrow bacteria\rightarrow 2Fe_2(SO_4)_{3(l)}+2H_2O_{(l)} \quad (17)$$

One-step bioleaching process

The entire bioleaching mechanism occurs in a single stage, in which the mother culture is cultivated in an aseptic flask as an inoculum. This inoculum is added to the E-waste-containing nutritional medium and cultured for a few days. Using iron as a source of energy, this progressively converts the copper from CuO (insoluble form) to Cu^{2+} ions (soluble form). This approach works not only for copper, but also for Zn, Ni, and Al. Heavy metals are released from this method, which may restrict the growth of microbes.[23]

Two-step bioleaching process

This method does not involve the direct use of microorganisms along with E-waste. Under aseptic circumstances, the required microorganism is cultivated in the appropriate culture medium. If the microbial community reaches the exponential phase, the waste sample is introduced as the second step. This step provides the optimum temperature for the growth of microorganisms and reduces the inhibition of rate. Thus, the two-step process is more appropriately used for the bioleaching process.[88] The microorganisms used in the bioleaching procedure and its findings are listed in the Table. 3.

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Table 3. Tabulation on bioleaching process and its findings.

Microorganism	Type of Bioleaching	Metals recovered	Findings	Reference
Acinetobacter species Cr B2	Pulsed plate rector	Copper	Extracellular enzymatic and metabolite function requires minimal energy, and are easily controlled	[89]
Leptosprillum ferriphilum, Acidithiobacillus caldus	Stirred Tank Reactor	Aluminium, Copper, Lead, Tin	Have better performance, heat resistant bacteria, used widely in the semi-industrial process	[90]
Acidithiobacillus ferrooxidans	One-step	Copper	Graphene as a catalyst for Cu bioleaching	[91]
Acidithiobacillusferrooxidans	One-step-Bioreactor	Copper	Carbon nanotube's synergetic effect is investigated	[92]
Leptosprillumferriphilum	Two-step	Copper, Tin, Chromium, Nickel, Zinc	Pyrite used as lixiviant	[93]
Acidithiobacillus ferroxidans	One-step	Copper, Lead, Nickel, Zinc	The citric acid (natural lemon juice)as a chelating agent improves the metal recovery	[94]
Acidithiobacillus (A.) caldus	One-step	Iron	Using biogenic H_2SO_4 for the production of biogenic Fe^{3+}	[95]

b. Indirect bacterial leaching

Indirect bacterial leaching is the process by which microorganisms create the lixiviant and solubilize the metal without coming into direct contact with the waste's base, as shown in the equation (18) and (19) below:

$$MeS_{(l)} + Fe_2(SO_4)_{3(l)} \rightarrow MeFeSO_{4(l)} + 2FeSO_{4(l)} + S^0_{(s)} \quad (18)$$

$$2S^0_{(s)} + 3O_{2(g)} + 2H_2O_{(l)} \rightarrow \text{bacteria} \rightarrow 2H_2SO_{4(l)} \quad (19)$$

Figure 7 depicts a schematic illustration of the direct and indirect bioleaching processes.

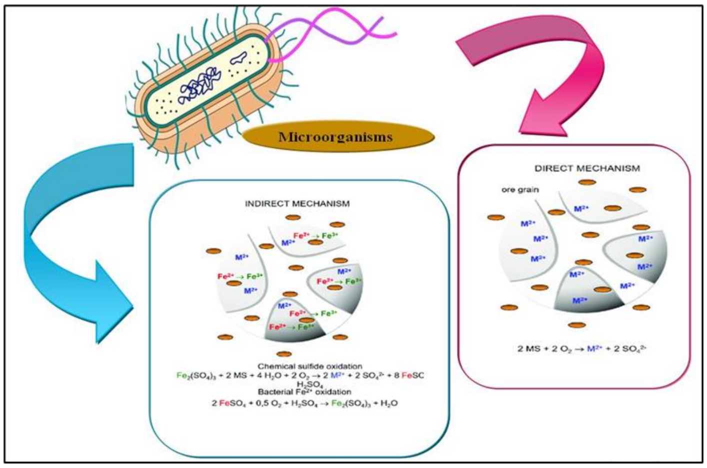

Fig. 7 Schematic representation of direct and indirect bioleaching process.

c. Spent medium process

This approach employs a two-step process for microbial culture in conjunction with livixants. The spent medium is collected from the microbial culture and combined with the E-waste sample. Because of its ability to minimize the toxicity of PCBs on the growth of microorganisms, this procedure is mostly employed for gold recovery when compared to other processes. According to a study conducted by East China University's State Key Laboratory of Bioreactor Engineering, bioleaching of PCB recovers 93.4% of copper in 9 days.[3,96] The alloy-resistant *Streptomyces albidofavus* TN10 isolated from a termite colony was studied by Kaliyaraj *et al*. for the valuable metal recovery from PCBs. They used the bacterial strains which were inoculated in the ISP medium with PCB powder at 28°C in a rotator shaker (110 rpm) for 120 hours. Under laboratory conditions, microorganisms were reported to recover heavy metals such as Zn 82%, Ni 81%, Cu 68%, Cd 65%, Ca 74%, Al66%, Ag 56%, Pb 46%, and Fe 42% within 72 hours. They were analyzed for the recovery metal concentrations at three different pH conditions. Maximum recovery occurs at pH 6 and its graphical representation is mentioned below in Figs. 8(a, b and c).[97]

Most of the studies conducted on the bioleaching of PCBs have been limited to simple laboratory-scale tests. However, as of the end of 2021, more comprehensive, larger-scale work has been carried out, revealing that: (1) indirect bioleaching can be applied at a commercial scale; (2) microbial consortia can be adapted to PCB leachate and raffinate; and (3) microbial colonization of PCB leaching reactors can be advantageous. Moreover, the biological oxidation of iron in PCBs can provide enough oxidant for the process to be continuous and the iron can be recycled. Nonetheless, some issues, such as acid consumption, still need to be addressed. Several studies are now scaling up the process, aiming to optimize the operating conditions, gather techno-economic and environmental data, and evaluate the commercial feasibility of PCB bioleaching.[98] On the other hand, Synthetic biology is a promising field that can be applied to biohydrometallurgy.[99] The use of microorganisms in this field can be restricted by inhibitory substances present in the ore or

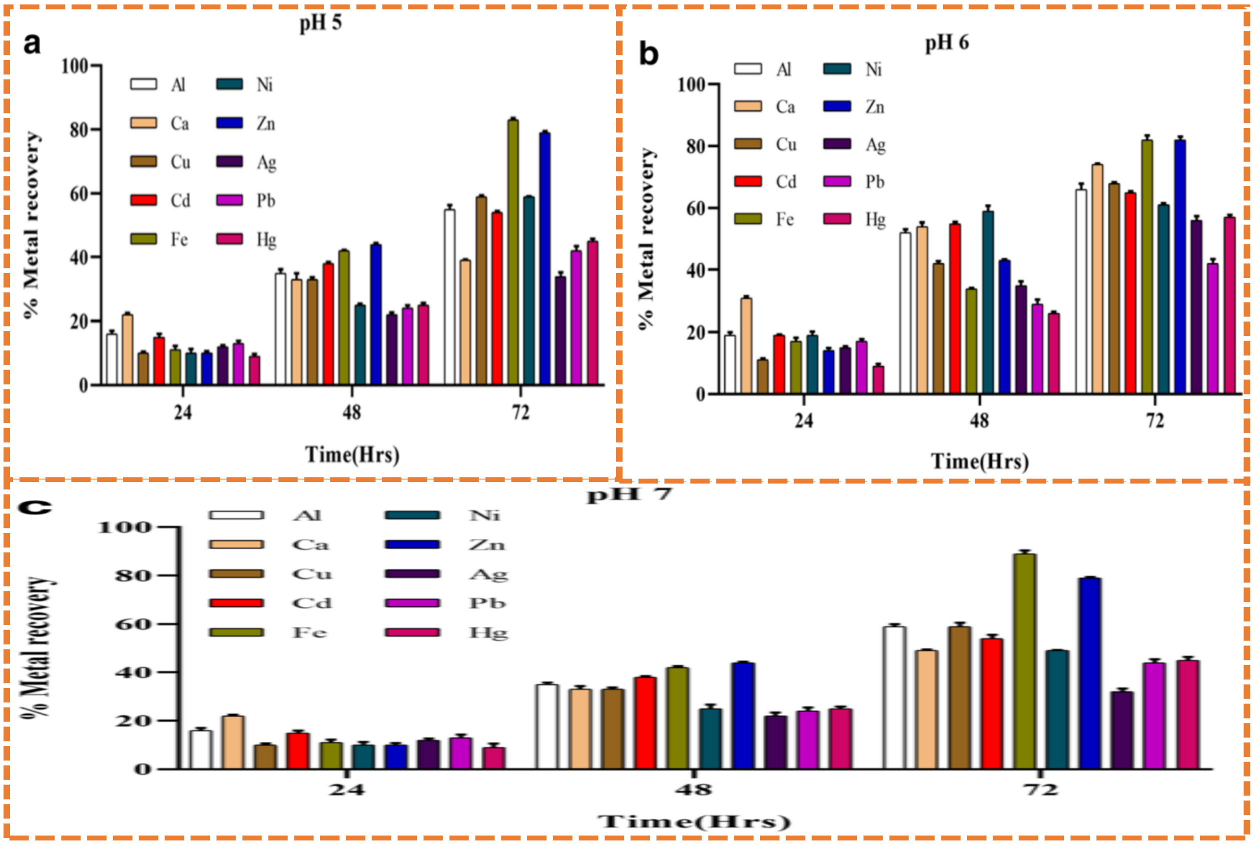

Fig. 8 Graphical representation of recovery of metals by bioleaching from PCBs at various pH(a:pH 5, b:pH 6, c:pH 7). Reproduced with permission from [97].

waste being treated. Synthetic biology, which involves the design and construction of new biological systems using engineering principles, offers potential solutions to overcome these limitations. One approach involves engineering microorganisms with resistance to stressors such as high metal concentrations, extreme temperatures, and high salt stress, with improved metabolic pathways of iron and sulfur oxidation. This approach has already been successful in improving gold bioleaching from electronic scrap using cyanogenic microorganisms. An engineered strain of *Chromobacterium violaceum*, which produces and detoxifies cyanide, was modified to contain an additional operon for cyanide production. This engineered strain demonstrated the highest cyanide production and, correspondingly, the highest gold recovery rate of 30% at a 0.5% w/v pulp density. This is a significant improvement over the wild strain, which enabled a mere 11% recovery of gold.[100]

3. Recent breakthroughs in metal recovery

3.1 Nanotechnology in Recycling e-waste

Nanomaterials with sizes ranging from a few nanometers to 100 nm have the potential to provide promising solutions to a broad range of ecological concerns. These nanomaterials can remediate waste more efficiently than their bulk counterparts because they are smaller in size, more flexible, surface-active, environmentally safe, and have unique physicochemical features. Furthermore, these materials can be customized because they can be employed as nanofilms, nanowires, nanocolloids, nanotubes, and nanoquantum dots. The topic of nanotechnology has attracted numerous researchers to employ them in a range of applications as the use of nanoscale materials becomes more widespread. The recovery of metals from e-waste at the nanoscale has been the subject of several scientific articles.[101,102] For example, a study[103] reported a low-temperature technique in which WPCBs were ball-milled into nanosized particles with improving downstream component recovery. Similarly, e-waste was utilized to recycle metals such as Cu, Au, Fe, Hg, and Pb which were then used to create nanoparticles. The green synthetic bottom-up generation of nano-sized CuO particles from bio-hydrometallurgical leachate was previously discussed by the authors.[104,105] The recovery of nanoscale materials through the recycling of electrical and electronic products has thus introduced a new phase in environmental research and nanotechnology.

3.2 Precious metal recovery via photocatalysis

Under modest conditions, photocatalysis has been shown to create a sequence of reactions that produces extremely reactive free radicals. Furthermore, photocatalytic technology has the advantages of being simple to use, consuming little energy, producing no secondary pollution, and being highly efficient. Because of its direct use of solar light-driven reactions and strong catalyst stability, photocatalytic oxidation

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has become a preferred technique for addressing environmental pollution and energy crises. The use of photocatalysis to dissolve valuable metals without the use of powerful acids, bases, or poisonous solvents also becomes one of its major advantages. It has a good leaching impact on seven different precious metals (Au, Ag, Pt, Pd, Rh, Ru, and Ir). For example, light irradiation successfully dissolved gold (Au) from a central processing unit (CPU) board and gold ore, as well as platinum (Pt), palladium (Pd), and rhodium (Rh) included in TWC. The reaction conditions required are light, and the raw materials are simply added and combined. Crushing the bulk samples increases the reaction contact surface, allowing more metals to dissolve. Several metals, including copper (Cu), nickel (Ni), and gold (Au), were dissolved from the CPU board. These non-noble metals can also be dissolved during the photocatalytic dissolution process. The photocatalytic procedure has a milder reaction than the aqua regia approach. Aqua regia dissolution is a violent reaction that produces a high number of toxic and dangerous chemicals, like chlorine. As a result, photocatalysis is a highly feasible new strategy for metal recovery.[106]

3.3 NBS and Py-based recovery

A novel chemical technique for metal recovery based on the simultaneous usage of N-bromosuccinimide (NBS) and pyridine (Py) with several lower environmental effects, reagent cost, and reaction conditions. As a result, it announced a unique NBS and Py-based gold extraction technique. This technique leached Au(0) from gold ore and electronic waste at room temperature at a pH that was almost neutral, transforming it to Au(III) with good yield and selectivity. This method is significant because it offers a simple, environmentally sustainable, and feasible option for gold leaching by reducing overall chemical waste and energy burden. It also displayed low environmental effects and cytotoxicity.[107]

3.4 Progress in Bioleaching

In recent years, laboratory-scale bioreactor experiments have led to the development of biohydrometallurgy for PCB processing, achieving over 90% copper recovery rates.[108] An example ofthis progress is the enhanced sulfide oxidation and gold recovery, with a reduction in energy consumption of at least 25% at an operating temperature of 38-40 °C. The bioreactor consortium called as BIONORD process is mainly composed of *Acidiferrobacter spp.* And *Ferroplasma spp. (over 50%), with Acidithiobacillus* and *Acidiphilium spp.* accounting for 26% of the population resulting in the production of 30 tonnes of gold, which accounted for approximately 0.83% of global gold production of the year.[109]

4. Application &future perspectives

OMICS uses a new technology in genetics, genomics, metabolomics, and proteomics as well method identifies microorganism properties related to their interactions with proteins, macromolecules, genes, and the environment. Genomics studies the structural organization, function, and evolution of the genome, metabolomics studies the molecular changes in cellular development, and proteomics studies the organization, function, and interaction of proteins. In addition to bioinformatics applications, the other OMICS technique contributes to the advanced knowledge of the physiology of microorganisms by participating in the bioleaching processing system.[110,111]

Tay *et al.* utilized metabolically modified pBAD and pTAC of C. violaceum strains for the retrieval of gold in their work. Both strains boost gold recovery; however, pBAD and pTAC produce 68% and 51% cyanide lixiviant, respectively, when compared to comparable non-engineered cyanogenic bacteria. Proteomics research has been used to investigate the modulations of the lixiviant metabolic network of pBAD[112]The OMICS data analysis approach was used to investigate the effect of factors on the diffusion of ions from mineral ores by bioleaching.[113] Natarajan and Ting investigated the mutation of alkali-tolerant bacteria (C.violaceum) by focusing on growing in highly alkaline conditions suitable for chemical stability to create cyanide. At pH 9.5, this mutant strain recovers a high amount of gold.[114] Thus, OMICS will be helpful for the determination of the exact mechanism behind the leaching of metals using the microbes in near future.

5. Conclusion

Several researchers are concerned about the environment and drive the practice of recycling valuable metals from e-waste with minimal environmental impact. These conventional techniques were widely performed by various researchers and industries for efficient metal recovery.

• In particular about the process of metal recovery, pyrometallurgy was generally used to extract metals from e-waste. However, some of the process in this method releases harmful gasses such as dioxin which acts as a major threat to the environment, and the human health and smelters used in this are highly dependent on high investments.

• Due to this drawback, hydrometallurgical techniques for recovering metals from electronic trash with less negative environmental effects were developed. Over 100 years cyanide leaching wasused by mining industries which paves the way for the recovery of metal with good economic feasibility. It is concluded the recovery of gold by thiourea is the most suitable method due to its rapid interaction with the gold as well as havingless impact on the environment when compared with other methods.

• In recent years, due to the advancement of biotechnological techniques, it has been widely used and achieved higher yields which arebeneficial for small as well as large-scale industries. Compared to the other conventional methods, the use of microbes for the leaching of metals offers minimization of the use of chemicals with low operating costs.

In conclusion, this review highlights the major methods

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and current trends in the recovery of metals from electronic waste. These methods include hydrometallurgy, pyrometallurgy, and biometallurgy, each with its advantages and limitations. In recent years, there has been a shift towards sustainable and environmentally friendly methods, such as biometallurgy and electrochemical methods. However, recovery of metals from e-waste is a crucial step in the circular economy which can help to reduce the environmental impact of electronic waste. So, additional research is needed to improve the efficiency and effectiveness of these methods and to develop new techniques for the recovery of valuable metals from e-waste.

Acknowledgment
This work is supported by DST-SERB, New Delhi, Project File number: EEQ/2021/000467EEQ.

Conflict of Interest
There is no conflict of interest.

Supporting Information
Not applicable.

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