

Review Article

Toxic Metal Recovery from Waste Printed Circuit Boards: A Review of Advanced Approaches for Sustainable Treatment Methodology

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The rapid advancement of technical advancements has resulted in the generation of substantial amount of electronic trash (e-waste). The volume of e-waste created, as well as the presence of both dangerous and beneficial elements, enhances the business potential of recovery and recycling significantly. Waste printed circuit boards (PCBs) include a number of hazardous heavy metals, including copper (Cu), tin (Sn), lead (Pb), and others (Zn, Ni, Fe, Br, Mn, Mg etc.). These discarded metals without treatment threaten the economy, the environment, and human health. Heavy metal recovery from PCBs is a big difficulty for researchers. The present review focuses on technological advances in the recovery of toxic, precious metals from PCBs.

1. Introduction

1.1. E→Waste Generation Scenario. Electronic garbage (e-waste) is becoming a major global problem due to rapid technological obsolescence and informal recycling and reuse techniques [1, 2]. Electronic equipment has a fixed lifespan and stop working after a certain time. These discarded items are repurposed after any necessary refurbishment. Products that can't be reused are recycled or thrown away in landfills as scrap. Due to the equipment's decreasing life-span and increased demand, this volume is projected to increase in the future [3, 4]. As a result, e-waste has become the world's fastest-growing rubbish volume, with an annual growth rate of 5% and a total volume of over 50 million tons by 2020. The issues that these compounds cause get more serious as they damage the ecosystem.

Over the previous two decades, the global market for e-waste has expanded significantly, while the lifespan of such devices has decreased. Many of the components in e-waste are dangerous and non-biodegradable, which causes

concern. Toxic metals including lead, copper, tin, aluminum, nickel, zinc, and other hazardous components are discovered in e-waste and are either burned or recycled, depending on the treatment process. Several toxic chemicals were emitted throughout the procedure, resulting in a range of environmental difficulties, human health consequences, and ecological damage. In 2019, just 17.4% of the world's 53.6 million metric tonnes of e-waste was recycled [5]. China generated the most e-waste, with 10.1 million tonnes, followed by the United States, with 6.9 million tonnes. India, with 3.2 million tonnes, was third. These three countries accounted for around 38% of worldwide e-waste. The steady growth in E-waste generation rates is due to the country's population and ongoing technological advancements.

1.2. Printed Circuit Boards. A printed circuit board (PCB) is an electrical circuit board that connects components. Due to characteristics like as complex structure, high metal content

and possible risks, discarded PCBs are known as the most challenging components of E-waste to recycle. A typical PCB is made up of 40% metals, 30% ceramics, and 30% plastic components [6–13]. Waste PCBs now account for 3–6 wt% of total e-waste created. On the one hand, discarded PCBs include heavy metal components, organic compounds, and chemical residues that are hazardous to the environment and human health. Waste PCBs, on the other hand, have a significant residual value since they include high-grade precious metals such as Au, Ag, Cu, Pd, and so on by about 28% weight. As a result, recycling discarded PCBs is vital for both environmental preservation and economic development. Figure 1 shows a sample PCB.

1.3. Components in PCBs. A typical PCB trash may include up to 60 unique components, some of which are useful while others are hazardous. PCBs comprise metals (Cu, Sn, Pb, Ag, Au, Pd, Fe, Ni, and Cr), nonmetals (glass fibers, electronic component insulators, capacitors, resistors, and so on), and organic compounds (epoxy resin, paints...). Copper is the primary component of PCB, which is employed as an electric current conductor. PCBs, particularly electronic PCBs, contain precious metals. Palladium is utilized in contacts and multilayer ceramic capacitors, whereas silver is used in solder and contacts, gold as a protective later on contacts. Table 1 shows the typical metallic concentration of PCBs based on literature [6–18]. In our previous studies [4], the principal components of the PCBs were analyzed using SEM and EDX analysis, as shown in Figures 2(a) and 2(b).

1.4. Environmental and Health Impacts of Toxic Metals of PCBs. The improper disposal of abandoned PCBs aggravates environmental problems while also harming human health. Toxic metals contained in PCBs discharged into water, air, or landfills stimulate the formation of micronuclei and chromosomal anomalies, resulting in genetic instability in individuals exposed [19, 20]. When hazardous substances reach the human body, they spread to a number of tissues and organs, where they are metabolized and can participate in a variety of physiological processes. Previous researchers reported the following toxic effects: Pb (affects reproductive, mental instability, cytotoxicity, ischemia and trauma, and damages human DNA) [21–27], Cu (headache, dizziness, irritation in eye, nose, mouth) [28–30], Sn (effects in central nervous system disorders and visual defects) [31], Ni (lung disfunction, asthma, skin allergy, carcinogenic effects) [30, 32–34], As (breathing issues, increased risk of blood). Figure 3 highlights the e-waste sources in various sectors and their effects on human health.

1.5. Need for Study. The extraordinary growth in the use of electronic products creates massive amounts of e-waste all around the world. Metals can enter the water system at any moment through industrial and consumer waste, releasing potentially hazardous heavy metals into streams, lakes, rivers, oceans, and groundwater [35, 36]. As a result, a reliable method of extracting metal from PCBs is required. The

selection of an appropriate recovery method will be critical to the successful treatment of PCBs. The primary research priority is the development of breakthrough technology for removing harmful heavy metal ions from discarded PCBs. The present state of PCBs treatment via pyrometallurgical and hydrometallurgical processing, as well as technical breakthroughs for sustainable waste PCBs recycling were described in this study.

2. Technologies for Recovery of the Metals

Many recycling approaches have been investigated in traditional operations, including pyrometallurgy, hydrometallurgy, and a combination of the two. In terms of price, environmental effect, and metal recovery, each process offers benefits and disadvantages. The uniqueness of effective treatment alternatives is determined by the choice of an appropriate recovery plan [37, 38]. The research and development of innovative heavy metal recovery technology is a major scientific undertaking. Several approaches used in previous research to extract harmful compounds from PCBs were addressed in this study.

2.1. PCBs Recovery through Pyrometallurgical Processing. The science and technique of extracting or refining non-ferrous metals from metallurgical materials at extreme temperatures is known as Pyrometallurgy. Pyrometallurgy of PCBs refers to the use of burning and other methods to remove nonmetallic components from the circuit board, allowing the metal to be enriched and recycled further. In comparison to hydrometallurgical technology, the use of pyrometallurgy technology to dispose of discarded circuit boards has a high processing volume and is a straightforward, easy-to-operate operation [39]. It primarily recovers metal components at a low cost and with high efficiency.

2.1.1. Incineration and Pyrolysis. The incineration of abandoned PCBs is a straightforward and effective method for accomplishing successful disposal and energy recovery. It inhibits the formation of melts rather than smelting. The approach is useful in the small-scale processing of electronic waste with a high concentration of valuable metals. The temperature of the smelting furnace is generally set above 1000°C to limit dioxin emissions. Pollutants from incineration harm the environment and stifle industrial development [40, 41]. The thermo chemical decomposition of organic material at high temperatures is known as pyrolysis. Pyrolysis recycles PCBs by converting the organic part into low molecular products (liquids or gases) and making the PCB brittle and easily crushed [42]. The pyrolysis of discarded PCBs inhibits the formation of dioxins since it is performed in the absence of oxygen. Although there has been a lot of study done on the pyrolysis of PCBs, the most of it has been done in nitrogen settings using analytical pyrolysis methods or laboratory size reactors with kinetics measurement and product characterization. Much research on incineration and pyrolysis has been undertaken, and it has been discovered that they cause a variety of

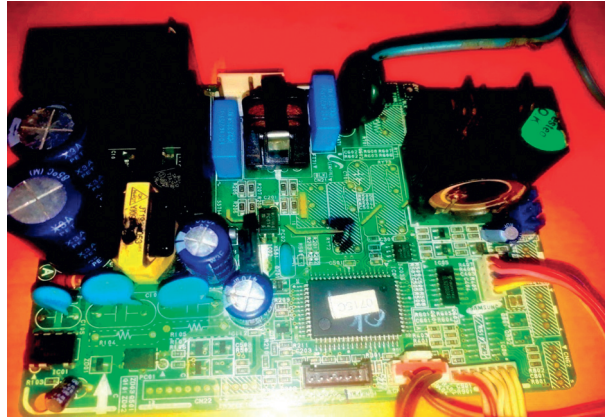


FIGURE 1: Schematic diagram of sample PCB.

TABLE 1: Metal composition present in printed circuit boards reported in previous literature.

Metals elements in PCBs	Metals presents in PCBs according to previous reports (Wt %)								
	[6] %	[7] %	[12] %	[13] %	[14] %	[15] %	[16] %	[17] %	[18] %
Pd				0.025	0.021	0.022		0.003	
Pb	3	2		4.19	0.3	2.96	2.50	1.3	
Al	7	2	4.7	4.78	1				
Sn		4	1			5.62	4.79	3.8	
Au	0.011	0.1	0.008	0.725	0.035	0.025	0.014	0.0068	
Mg									0.12
Ni	2	2	1.5	0.95	0.1	1.65	0.41		0.61
Cr						0.356			
Zn	0.4	1	0.45	2.17			0.18		
Sb		4	0.06				0.05		
Cd							0.1183		
Cu	24	20	26.8	6.5	13	14.6	14.2	28.7	24.2
Ag	0.028		0.33	0.223	0.134	0.045		0.0079	
Fe	12	8	5.3	0.11	5	4.79	3.08	0.6	0.18

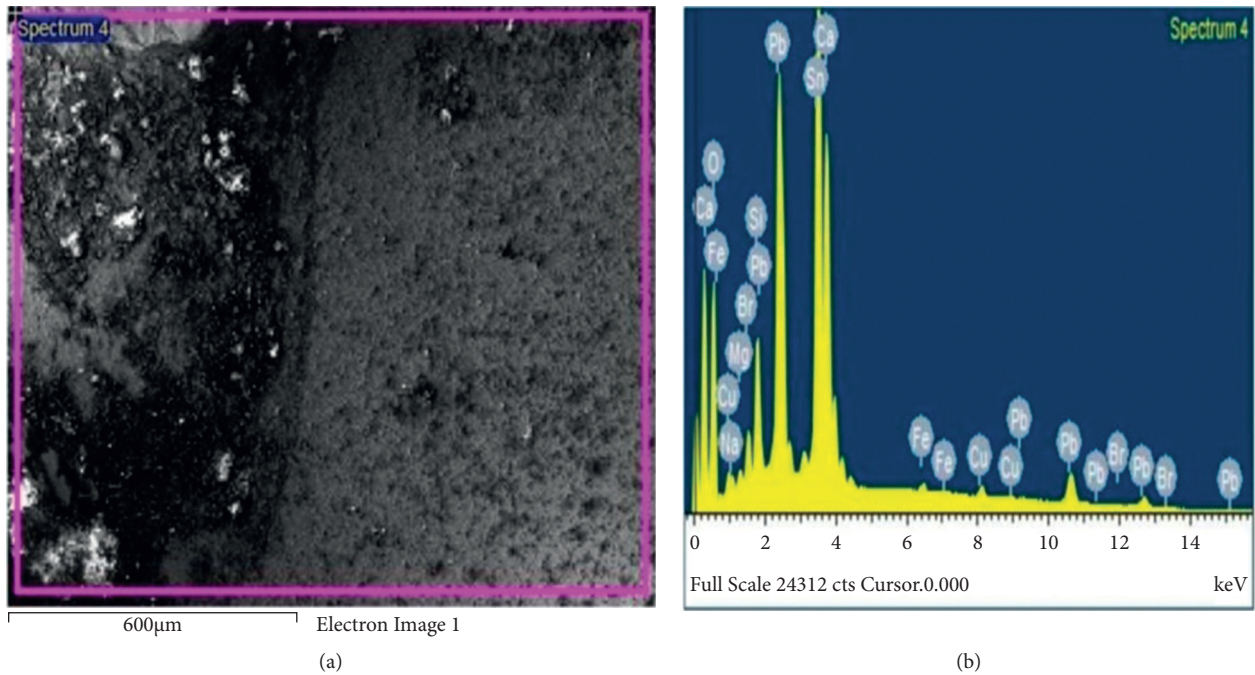


FIGURE 2: (a) SEM spectrum analysis of sample PCBs, (b) EDXs spectrum analysis.

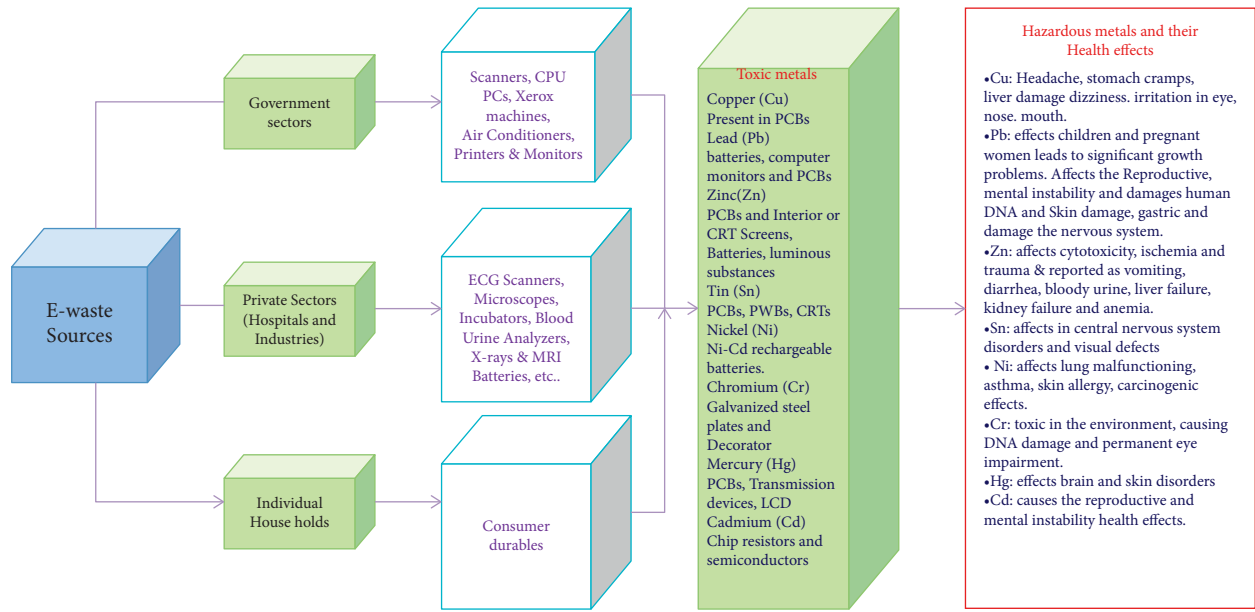


FIGURE 3: Schematic diagram of E waste sources in various sectors, and their effects on human health.

environmental concerns, including global warming, as well as health impacts due to carbon monoxide and sulphur dioxide emissions. At the moment, the two practicable options to handle the nonmetallic component physically removed from PCBs on a wide scale are incineration and landfill.

2.2. Hydrometallurgical Process. When compared to pyrometallurgy, the hydrometallurgical process has lately been investigated due to its advantages, which include low capital costs, less environmental impact, and simplicity of management. Metal leaching, purification, and recovery are the main parts of the hydrometallurgical process.

2.2.1. Chemical Leaching. Chemical leaching, which involves dissolving heavy metal ions in a leaching media, is a common approach for removing heavy metal ions from PCBs. Several investigations have been conducted on the extraction of metals from PCB waste. Because of their potency and low reagent cost, various inorganic acids, such as H_2SO_4 , are used in the literature. Inorganic acid leaching, on the other hand, uses more water and chemicals while producing secondary waste. Previous studies employed a range of chemical reagents to extract metals from PCBs, including nitric acid, hydrochloric acid, sulfuric acid, cyanide, ammonia, thiosulfate, hydrogen peroxide, ammonium per sulfate, and aqua regia. Leaching is performed in a one-of-a-kind ultrasonically aided treatment technique, followed by reduction, recovery, and separation. Copper and iron were entirely recovered by separating PCB waste sludge into copper sulphate and ferric chloride solutions [43]. To extract metals from PCB waste sludge, the technique has a high separation and recovery efficiency. This technique's efficiency for a metal recovery plant handling PCB waste sludge comprising 3.14–4.85% copper and 3.7–4.23% iron yields a copper recovery efficiency

of 95.2–97.5% and an iron recovery efficiency of 97.1–98.5%. 10 g of sample and 50 ml of distilled water were utilized in the supercritical water oxidation (SCWO) treatment studies, with hydrogen peroxide as a source. For various time durations, the experiment was carried out in a 200 ml high-pressure reactor. Within 11 hrs of treatment, about 84.2% of copper was recovered in the cathode compartment, of which 74% was deposited on the cathode with a purity of 97.6% and may be immediately reused [44]. Furthermore, [42] showed effectively that copper was leached by six ionic liquid (IL) acids. The concentration of IL acid, quantity of hydrogen peroxide, solid to liquid ratio, and temperature all have varied impacts on copper leaching from waste PCB. When IL acid is mixed with sulfuric acid, copper is considerably simpler to leach out of discarded PCB powders. In this case, IL acid diffusion is critical to the surface reaction, whereas copper leaching by inorganic acids normally regulates the surface reaction. It was given increased environmental impacts owing to the usage of more harmful acids.

The use of acidic ferric chloride solutions in electrochemical oxidation proved to be an efficient way for simultaneously recovering copper and separating gold-rich residue from discarded PCB. The experimental results show that the longer length of the scaling process may improve copper extraction performance and raise the gold content of the solid residue to a greater extent. A laboratory-scale leaching unit was built, allowing 99.04% recovery of a high purity copper deposit [45]. The chemical leaching of copper from abandoned refrigerators was investigated using several techniques that were optimal in different scenarios. Studies were performed on the removal of Cu to identify the optimum disposal conditions for E-waste given the constraints of sound environmental handling using decision analysis tools [46]. Research [47] has tested the effect of hydrogen peroxide on sulfuric acid leaching of zinc, copper, iron, aluminum, and nickel.

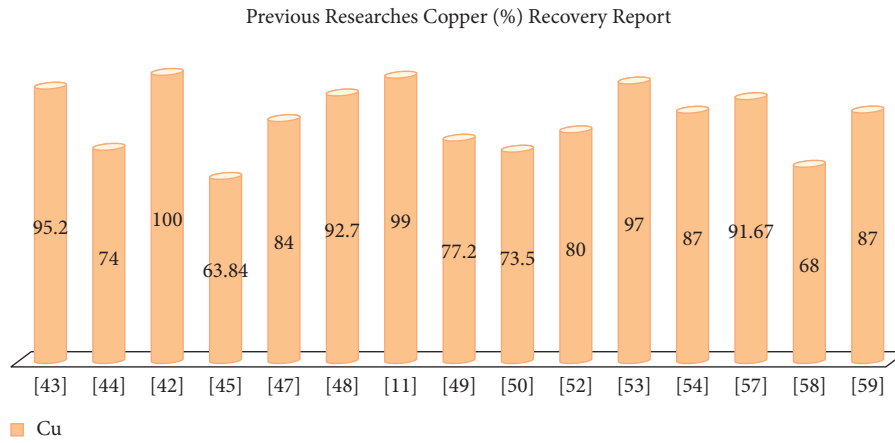


FIGURE 4: Graphical representation of copper recovery from previous studies.

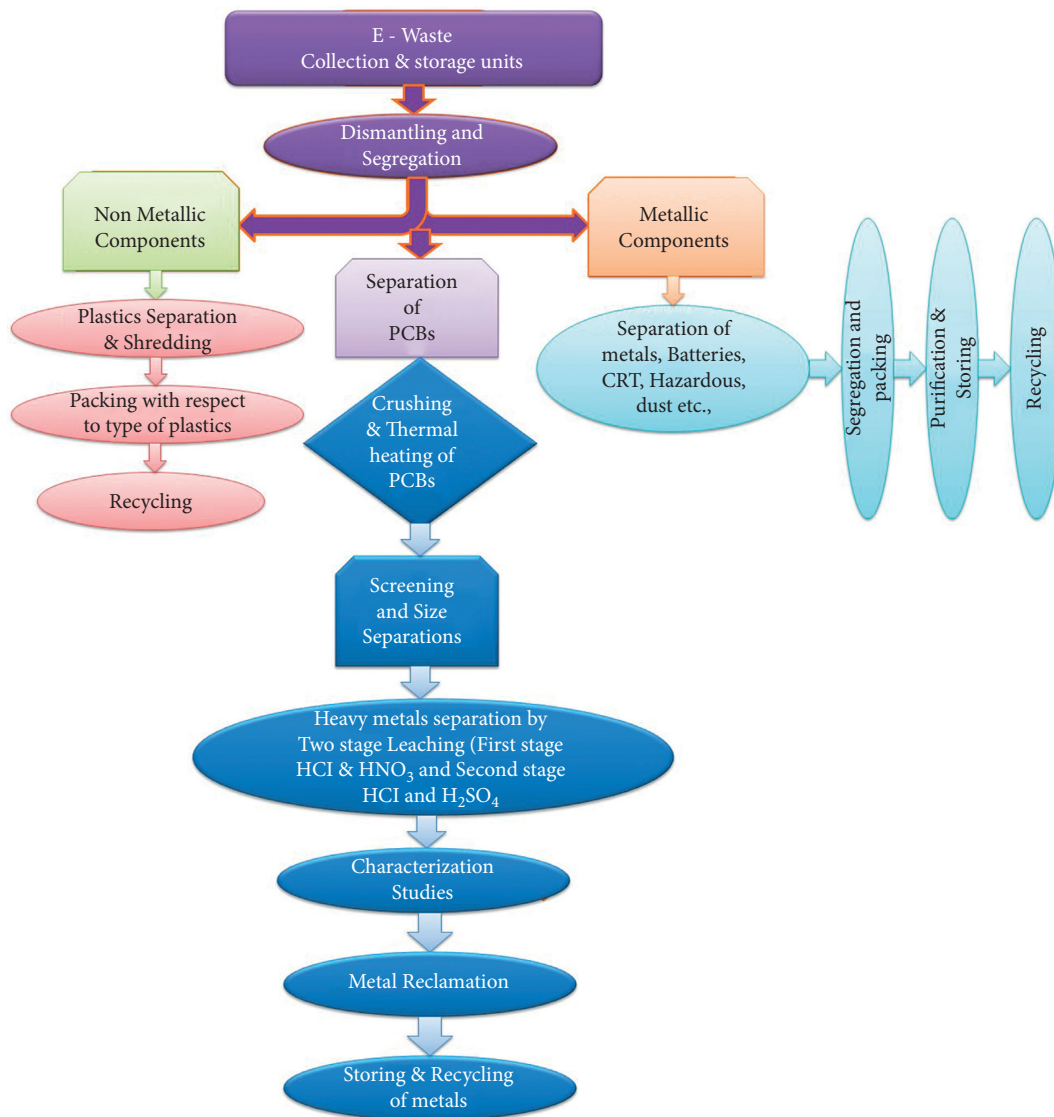


FIGURE 5: Flow diagram for metal treatment and recovery, and recycling.

The recovery of gold and silver from PCB waste was studied using ammonium thiosulphate in the presence of cupric sulphate and ammonia. Leaching with sodium chloride was used to assess lead and tin recovery. With some waste unrecoverable, the overall efficiency of recovered metals was 84% Cu, 82.1% Fe, 77.6% Al, 76.6% Zn, 70% Ni, 90% Pd, and 88.6% Pb. The research examined the developing issue of e-waste, which is being driven by the fast expanding volume of complicated end-to-end life of electronic equipment. There are significant fluxes of both harmful and useful compounds at the global level of production, consumption, and recycling. Despite the fact that knowledge and preparedness for implementation and improvement are quickly rising, there are several barriers to managing end-of-life items safely and efficiently in industrializing countries. Support ensures the economic and long-term viability of the e-waste management system by increasing the value-added and enhancing the collection and recycling system's efficacy. The quantity of copper removed in the study's leaching system was 92.7%; the precipitate produced by neutralizing the leach liquid was 9.77% [48]. For PCB removal, chemical leaching has been used, in which the PCBs are broken using a crusher and sieved with a screener. The metal content was evaluated by AAS by dissolving it in aqua regia. The study reported that at optimum conditions, about 99.9% of copper was leached with 60.0% of zinc, 9.0% of nickel, and a nondetectable amount of iron. At the cathode, 99.97% pure copper was obtained after electro-winning the leached solution. From this study, it could be concluded that the metal leaching from waste PCB in an alkaline solution is feasible [11].

The study investigated and reported on the various methods and conditions utilized to recover heavy metals found in PCBs, such as gold, silver, nickel, and copper. PCBs contain 80% precious metals in particle sizes ranging from 3.33 mm to 0.43 mm. According to column leaching data, the gold dissolving rate is higher than that of silver and copper during the first 10 days of the method. On day 11, gold and silver recovery rates began to fall due to a reduction in precious metal contact area caused by copper oxide and copper hydroxide layers on the material surface. In a column, cyanidation of PCBs yielded recoveries of 47.9% Au, 51.6% Ag, and 77.2% Cu [49]. To recover tin and copper, an 18% nitric acid solution was created, and 500 g of PCB were dissolved in it for 2 hours, until all of the solder was dissolved. The PCB components were removed, and the solution was filtered using filter paper to get copper nitrate and stannic acid in the precipitate. It was cooked for an hour in the muffle furnace at 600°C to produce stannic oxide. When the concentration of nitric acid was lowered, the leaching time rose dramatically. For more than 50% concentrated acid, the reaction was immediate and completed in 15 minutes. When the acid concentration was less than 20%, the process took around two hours to complete. Copper was recovered with 73.5% efficiency by cementation from a copper sulfate solution [50].

In the experiment [51], 15 g granules of the mobile PCB sample were leached in a 250 ml solution using a 500 ml glass beaker containing the predetermined quantity of

ammonium thiosulfate and copper sulfate at different pH levels. All leaching studies were carried out at a speed of 250 rpm for the agitation. After 8 hours of leaching, the solution was withdrawn and filtered using Whatman 40 filter paper to remove the remaining PCBs. The residue was then dried in a vacuum oven at 130°C for 2 hours to eliminate all moisture from the sample. The samples were weighed, and the residue weight was computed. Under ideal circumstances, which comprised 0.1 M ammonium thiosulfate, a stirring speed of 250 rpm, and an 8 hr time period at room temperature, 56.7% gold could be leached from PCB granules. At thiosulfate 0.1 M, copper sulfate 40 mM, P^H 10–10.5, and a stirring speed of 250 rpm at room temperature for an 8 hr time period, the highest gold leaching was 78.8%. The cementation process by PCBs can create a solution and change the pH of the solution. They are then washed and filtered to eliminate the waste particles, yielding a pure copper tri-hydroxyl chloride filtrate. They begin the dissolve method by introducing hydrochloric acid. After being acidified with strong sulphuric acid, they react and convert to copper hydroxyl chloride, resulting in a copper sulphate solution. The final stage is the evaporation stage, in which the moisture is evaporated and the ultrafine copper sulphate crystals with 80% purity are obtained [52].

The copper was recovered using selective chemical leaching with sulphuric acid in the tests. The recovered PCBs are then mechanically crushed into a mesh size of 200. They are treated under working circumstances with 0.1 M sulfuric acid at pH 2.0 to 8.0 and temperatures ranging from 40 to 800°C. They were then analyzed using an atomic absorption spectrometer, and 97% of the copper was recovered [53]. Copper and tin were removed from approximately 73 kg of printed circuit boards using a heat treatment and a leaching process [54]. The trials were carried out in order to remove copper from printed circuit boards. They gathered different sized PCBs and evaluated their sizes using a filter mesh plate before dissolving the PCBs in acidic solutions. The copper is recovered with the greatest efficiency using sodium thiosulphate at a temperature of 200°C and leach durations of 10, 30, 60, and 120 minutes [55].

For the selective separation of electronic printed circuit boards, the study used a macro porous ion-exchange approach. The printed circuit boards are crushed to the appropriate size before being dissolved in nitric acid. They are then leached for 60 min at 25°C. The solution is then filtered. Cu is removed in 68.6% of the cases, Zn is removed in 56% of the cases, Ni is removed in 79.1% of the cases, and Fe is removed in 89.6% of the cases [56]. In the leaching studies of discarded printed circuit boards, sulphuric acid was employed to recover the valuable components of the sample after the PCBs were broken into tiny pieces and roasted for 1 hour at 600°C. Copper recovery is 87.6%, tin recovery is 94.0%, zinc recovery is 95%, nickel recovery is 81%, and Fe recovery is 58% [57]. For the cleaner manufacture of gold from secondary waste created during the leaching of base metals from PCBs, a novel recycling strategy based on electro- and solvo-chemical processes has been devised. In 0.2 mol/L solutions of thiosulfate and thiourea, respectively, 99 and 94% of the gold was recovered [58]. The copper

recovery is highly focused, and some of the process results are depicted in Figure 4. In summary, Figure 5 represents the overall set of operations for metal recovery, and recycling.

2.3. Bioleaching. High reagent consumption, secondary pollution, and high energy requirements drive research interest in the development of microorganism-driven recycling technology (bio metallurgy), which has the potential to be one of the most promising technologies in terms of low capital investment, labour-intensiveness, and energy consumption [59–63]. In bioleaching, bacteria and metals interact via reduction, oxidation, sorption, and sulphate precipitation. Historically, bioleaching has been utilized in industrial applications to recover metals from ores by bacterial leaching. The leaching method was demonstrated to be extremely successful using the bacterial strains *Acidithiobacillus ferrooxidans*, *Thiobacillus thiooxidans*, and *Acidithiobacillus*. These treatments were particularly beneficial since they were both environmentally friendly and cost-effective. Chemolithotrophic, heterotrophic, and thermophilic bacteria, as well as fungus, have all been examined. Commercial scale-up is problematic because the process is yet to meet the chemical leaching yields. To satisfy the process scaling requirements, genetically modified and mutant strains can be introduced [63]. Microbial growth broths, cyanide capture and enrichment reactors, and leaching reactors can all be separated to maximize lixiviant concentration and process pulp density.

3. Future Perspectives

The current leaching processes are inadequate for extracting critical metals. Future research should focus on reducing energy use and the cost of the recovery process. The combination of processes might result in a more efficient approach of extracting precious metals. A novel method for recycling strategic critical metals (those required to aid in the transition to green energy) from discarded printed circuit boards is proposed [59]. It is based on the combination of microbial activity with solvo- and electrochemical reactions in the process. Recent advancements in process integration for primary and secondary resource processing have brought promising outcomes. So far, however, no attempt has been made to analyze the variables and basics involved in process integration, which will benefit the complicated metallurgy of various important and vital resources [59–62].

4. Conclusions

Several studies on the recovery of heavy metals from printed circuit board waste using hydrometallurgical techniques have been published. The process's sluggishness and lengthier processing times were revealed to be limitations, resulting in less efficient recovery and a considerable impact on the recycling economy. Several research have been conducted to create more cost-effective methods of hydrometallurgical leaching followed by selective metal extraction. Earlier chemical leaching techniques, when compared to two-step leaching, required huge expenditures

in leaching reagents, high temperatures, pressure, and operations to be practicable for severe environmental problems. Bioleaching is yet to mature as technology for commercial usage. There is an ample scope in scale up of bioleaching process, which suffers with longer times, low yields.

Nomenclature

AAS:	Atomic adsorption spectroscopy
EDXs:	Energy-dispersive x-ray spectroscopy
EEE:	Electrical and electronic equipment
IL:	Ionic liquid
PCBs:	Printed circuit boards
PCs:	Personal computers
RSM:	Response surface methodology
SCWO:	Supercritical water oxidation
SEM:	Scanning electron microscopy
WEEE:	Waste of electrical and electronic equipment
WPCB:	Waste printed circuit boards.

Data Availability

All the data are available within the manuscript.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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