

Hydrometallurgical Recovery of Copper from Computer-Printed Circuit Boards

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Abstract. Modern electronic devices contain up to 60 different metals, leading to a growing interest in exploring secondary sources, particularly electronic waste (e-waste), to address potential shortages. This study focused on extracting copper (Cu) and gold (Au) from three different sources of computer-printed circuit boards (PCBs): board, USB jack, and capacitor. Aqua regia, a mixture of 68% concentrated nitric acid and 37% concentrated hydrochloric acid in a 1:3 ratio ($\text{HNO}_3=1:3$), was used as a leachant under various conditions, including different types of e-waste components, leaching times, and surface areas. Acidic leaching with a 1% (w/v) e-waste pulp density resulted in higher Cu extraction from PCB board waste compared to USB jack and capacitor waste. The optimal recovery of Cu achieved was 2207 mg/L after 20 minutes of leaching, using a particle size of 10 mm, at 100°C, 150 rpm, and a pulp density of 1% (w/v). The dissolution of Au was only traceable in the capacitor and USB jack with significantly lower quantities of 0.4 and 3.2 mg/L, respectively. Additionally, the electrowinning process managed to recover Cu with just 31.6% purity after one hour of electrolysis. In conclusion, this study demonstrates the feasibility of recovering valuable metals from e-waste through acid leaching, without the need for additional chemical treatments.

1 Introduction

The amount of waste electronic and electrical equipment (WEEE) has increased significantly in recent decades. The growth of the electronic and electrical industries, along with the increasing adoption of electronic technologies, has led to the creation of a larger number of new electronic devices [1]. As these products reach the end of their useful life, a significant amount of electronic waste (e-waste) builds up. In 2019, it was estimated that over 50 million metric tons of e-waste were generated, with projections indicating an increase to 74 million metric tons by 2030 [2]. The rapid and improper disposal of electronic waste has become a major concern, posing significant risks to both the environment and public health [3]. Furthermore, the incineration process has had detrimental effects on environmental quality, particularly concerning soil, air, and solid waste. The disposal of acid and sludge resulting from the burning of computer chips into the ground has led to soil acidification, which, in turn, contaminates water sources. Even more concerning, previous research has identified a range of health issues stemming from the toxic metals and chemicals present in e-waste. Chauhan's review confirmed the harmful impacts of metals such as lead (Pb), cadmium (Cd),

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mercury (Hg), chromium (Cr), arsenic (As), nickel (Ni), selenium (Se), lithium (Li), and americium (Am) on the human body [4]. Due to its potential health impacts, e-waste is classified as scheduled waste in Malaysia under Code SW110 of the First Schedule of the Environmental Quality (Scheduled Wastes) Regulation 2005 [5]. Code SW110 refers to WEEE that contains components like accumulators, mercury switches, glass from cathode-ray tubes and other activated glass, or polychlorinated biphenyl capacitors [5].

Even though contains over a thousand elements including toxic substances, e-waste includes non-ferrous metals like aluminum (Al), Cu and precious metals such as Au, silver (Ag), and platinum (Pt) [6]. To tackle these challenges, the concept of urban mining is emerging to advance the circular economy and encourage the recycling of metals found in electrical and electronic devices. This emerging practice has effectively turned discarded WEEE into a valuable secondary metal source [7]. Conventional techniques employed in the mining sector, such as pyrometallurgy, hydrometallurgy, and even bioleaching, can be applied to recover metals from these secondary resources. Pyrometallurgy encompasses processes and methodologies, such as incineration, smelting, and pyrolysis, which employ elevated temperatures to extract metals. Hydrometallurgy pertains to techniques that employ a chemical solvent to dissolve solid metals into an aqueous solution [8]. On the other hand, combining chemical and biological methods can improve the effectiveness of metal recovery from e-waste and reduce environmental problems [9]. Numerous researchers have suggested a combination of advanced metallurgical techniques and preliminary mechanical processing to manage the disposal of intricate WEEE scrap environmentally and economically [7].

To recover metals in their pure elemental form for reuse in new products, several methods have been suggested, including cementation, sulfide precipitation, ion exchange, and electrowinning. Electrowinning is a process involving the electrolysis of metal ions, which is employed after the leaching step to recover these ions via electrodeposition. Electrowinning has been successfully industrialised and can directly recover metals from acidic solutions, such as HNO₃, Aqua Regia, sulfuric acid (H₂SO₄) and HCl, as well as from ammoniacal solutions [10]. Among these, the electrowinning process is anticipated to yield the highest purity of the final product in the form of Cu foil [11]. However, the diverse metal content of PCBs complicates the electrowinning process, as the resulting leachate mixture from the leached waste contains various other base metals and components in addition to Cu [12]. Other issues like pH sensitivity, re-dissolution of metals, and overpotential are encountered in it [10].

This study aimed to recover two metals, namely Cu and Au, from three different components of computer PCBs. First, the metals were extracted using hydrometallurgy to leach the dissolved solid metals into an aqueous solution. Then, the electrowinning process was used to recover the metals in powder form. The effects of factors such as particle size and leaching time on metal recovery were investigated.

2 Materials and Methods

2.1 E-waste Sample Preparation

The e-waste, with a specific focus on computer PCBs, is sourced from Tess-Amm (Malaysia) Sdn. Bhd., an electronic facility located in Pasir Gudang Johor, Malaysia. Before transportation to the laboratory, no physical or mechanical methods are utilised on the sample to prevent any loss of its properties. The scrap computer PCBs (SW110) undergo thorough washing and cleaning to remove dust and any other adhering deposits. Two crucial stages of sample preparation preceding aqua regia leaching involve particle size reduction and component segregation for base metals recovery, particularly Cu. Initially, the computer

PCBs are manually disassembled using pliers and stainless-steel scissors into three distinct parts including capacitors, a USB jack, and the PCB board. Each part is then separated into individual 250 ml beakers. Subsequently, for the particle size study, the scrap is manually crushed into two different sizes with uniformity at 50 and 10 mm, respectively. Figure 1 depicts the computer PCBs both before and after segregation has occurred.

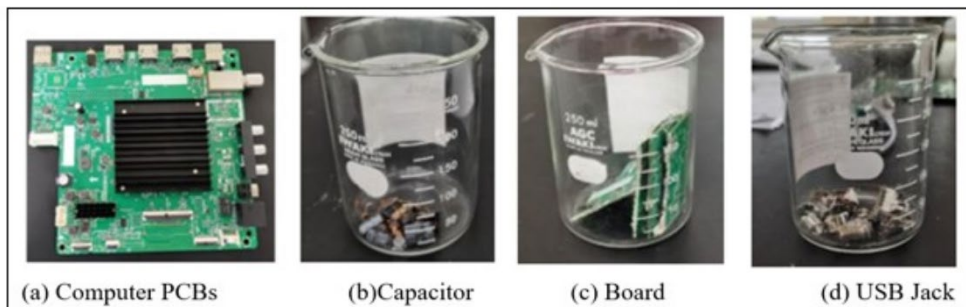


Fig.1. Scrap of computer PCBs and after manual segregation.

2.2 Aqua Regia Leaching

In experiments involving the aqua regia leaching of computer PCBs, a 250 mL beaker is employed. To prepare each experiment, a total of 100 mL of fresh aqua regia leaching solution is prepared by combining 68% concentrated HNO_3 and 37% concentrated HCl in a ratio of 1:3 (HNO_3 : HCl). All solutions are made using deionised water. The leaching experiments are designed to investigate the impacts of particle size reduction (10 and 50 mm), leaching time (20-60 minutes), and the influence of different components on Cu and Au leaching efficiency. The temperature during the experiment is maintained at 100°C with agitation at 150 rpm using a shaker. Liquid samples of 10 mL taken from each leaching experiment are analysed using an inductively coupled plasma-optical emission spectroscopy (ICP-OES) instrument, Model Optima 8000 from PerkinElmer, to determine the concentration of metal content in each sample. The samples undergo a 100x dilution for ICP-OES analysis.

2.3 Electrowinning Method

The recovery of Cu through the electrowinning process was analysed using a 500 mL glass beaker. Stainless steel and copper rods, each measuring 20 cm in height and 2 cm in width, were employed as anode and cathode, respectively. To ensure a clean surface free of passive oxide layers, the electrodes were pre-rubbed and rinsed before each experiment. The electrodes were weighed before and after each test. A Fisher magnetic stirrer was used to enhance the mass transport of the solution through agitation, and an AC/DC universal adapter provided the necessary current and voltage. Scale diagrams of the electrowinning setup are shown in Figure 2. The solution and the solid product formed at the cathode were separated using filter paper, dried in an oven and characterised. The reduction concentration of Cu metal ions was measured using an ICP-OES machine. The physical characteristics of the solid crystals were examined using Scanning Electron Microscope (SEM) analysis.

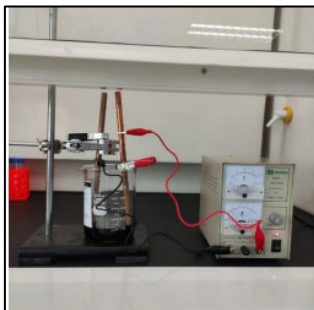


Fig.2. Electrowinning setup process.

3 Results and Discussion

3.1 Part of E-waste PCBs

Different types of e-waste components from computer PCBs were used in this initial study. Figure 3 shows that the dissolution of Cu increased consistently from the capacitor to the USB jack and board, despite using the same conditions: e-waste pulp density of 1% (w/v), 150 rpm agitation speed, and 100°C temperature. The concentration of Cu was highest in the board, USB jack, and capacitor samples, with contents of 6011.0, 2624.0, and 900.3 mg/L, respectively. However, for the precious metal Au, the amount of dissolution was only traceable in the capacitor and USB jack, with significantly lower quantities of 0.4 and 3.2 mg/L, respectively.

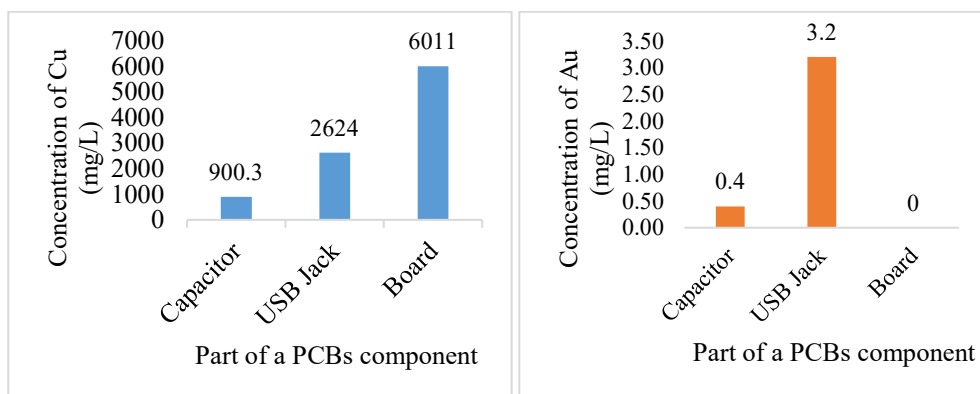


Fig. 3. Recovery of Cu and Au from different parts of e-waste PCBs.

The variations in metal concentrations can be attributed to the heterogeneity and complexity of e-waste [13]. The diversity in metal content within e-waste is influenced by its origin and type. Based on the results, the recovery of metals from 1% (w/v) of computer PCB boards using aqua regia was highest compared to capacitors and USB jacks. Therefore, PCB boards were used in the following experiments.

3.2 Effect of Particle Size

The second leaching experiment was conducted using the most effective part of computer PCBs: the board. This study focused on particle size to achieve optimal Cu recovery. Maintaining the correct pulp density (1% w/v) at an appropriate rate can enhance the available surface area relative to the amount of reagent utilised, thereby enabling greater Cu dissolution within a specified timeframe [14]. The operating conditions of temperature, agitation speed, and pH were kept constant, as these parameters were found to have no significant impact on Cu recovery efficiency. Therefore, the optimal conditions were determined to be 150 rpm at 100°C and 1% (w/v) pulp density.

Figure 4 shows the effect of particle size on the acid-leaching process for 20, 40, and 60 minutes of leaching time. The results revealed that optimal extractions of 2410 mg/L and 2185 mg/L of Cu were obtained after 60 minutes using board particle sizes of 10 mm and 50 mm, respectively. When the leaching time decreased to 40 minutes, Cu recovery performance slightly decreased, with a difference of 3.9% (loss of 95 mg/L) at 10 mm and 4.0% (loss of 88 mg/L) at 50 mm. However, there was a minimal difference in Cu dissolution during the first 20 minutes compared to after 60 minutes of leaching for both particle sizes, with only 8.4% and 8.7% differences using 10 mm and 50 mm particle sizes, respectively.

Even though the highest recovery of Cu was obtained after 60 minutes using a 10 mm particle size, from an economic perspective, the optimum leaching period is 20 minutes, as only a small difference is observed when operating for longer durations in the leaching process [15]. [16] reported that Cu is present in the middle layer of waste PCBs. When the particle size is large, Cu remains covered with the organic matter layer, resulting in poor liberation and low leaching efficiency. However, with a decrease in particle size, the degree of liberation between metal and nonmetal materials improves, enhancing contact between the metal and leaching solution and promoting copper leaching [16-17].

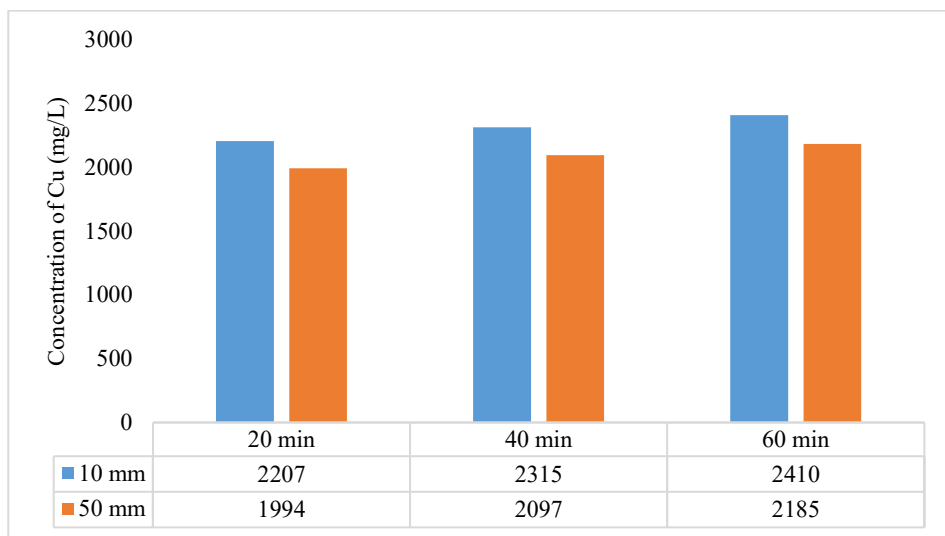


Fig. 4. Effect of board’s particle size towards the Cu recovery.

3.3 Electrowinning Results

The Cu leachate obtained from the highest recovery of 10 mm (2410 mg/L) was analysed using ICP-OES and subsequently used for electrowinning. Maximum Cu deposition, with a purity of 31.6% (difference of 761.6 mg/L) was achieved within an hour of electrolysis. The

experiment was conducted at a room temperature of 27°C, with a constant stirrer speed of 150 rpm using a magnetic stirrer to enhance metal extraction and deposition. A fixed voltage of 1V was applied throughout the experimental process, with currents increasing from 0.1 to 0.5A (uncontrolled). These results indicate a positive correlation between current density and metal deposition rate, suggesting that the process depends on both the period and the applied voltage [18].

Few previous studies can be directly compared to these findings, as most electrowinning research has used natural mining resources or metal compounds rather than e-waste sources. A study by [19] indicated that 91.1% of Cu could be recovered from an aqua regia solution containing Motherboard PCBs, using Al foil and a Cu rod as the anode and cathode, respectively. [18] employed the electrowinning method following a 2-step bioleaching process using acidophilic bacteria after metal dissolution. From their study, a 64.5% purity of Cu was recovered using a platinum rod and carbon felt as the anode and cathode. [20] reported a 99.6% purity of Cu from e-waste sources using an ammonium sulfate solution. While previous studies achieved Cu purities of up to 90%, the current study only attained a 31.6% purity. This result is believed to be due to the high pH of the aqua regia solution (pH below 2.0), as no pH adjustment was made before the experiment. Additionally, the use of a different anode type for the dissolution method may have reduced deposition recovery performance. The overpotential of the aqua regia solution could also have contributed to the lower recovery performance observed in this study [6]. With this, the findings of previous studies through the electrowinning method are included in Table 1.

Table 1. Deposition of Cu through electrowinning method by previous studies.

Type of e-waste	Purity of Cu (%w/w)	Current (A)	Voltage (V)	Anode	Cathode	Dissolution Method	Reference
Computer board (PCBs)	31.6	0.1-0.5	1	S/S rod	Cu rod	Aqua regia	This study
E-waste	64.5	0.05-0.2	2.8-6.1	Platinum rod	Carbon felt	2-step bioleaching	[18]
Motherboard PCBs	91.1	N/A	N/A	Al foil	Cu rod	Aqua regia	[19]
E-waste	99.6	N/A	0.0-2.5	Ti basket	Cu plate	Ammonium sulfate	[20]

* Remark: N/A: not available

The surface morphology of the recovered Cu after the electrowinning process, observed at 3000x magnification, is shown in Figure 5. The results illustrate the structure of the Cu molecules obtained during this experiment, consistent with previous studies using Cu rods as both cathode and anode [21].

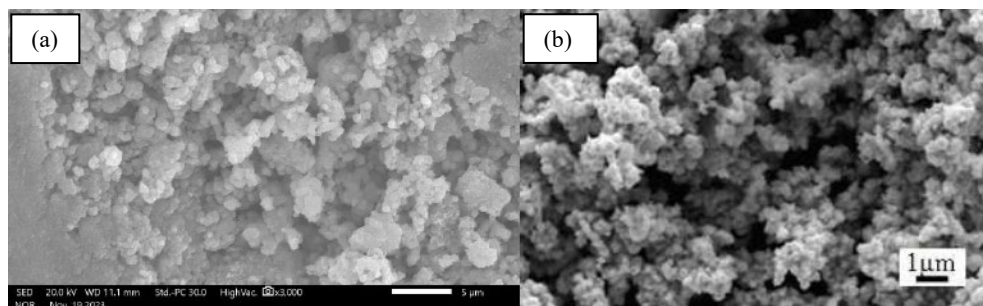


Fig. 5. SEM images of Cu deposition for (a) this study and (b) the study done by [21].

4 Conclusion

Three different parts of computer PCBs—capacitors, USB jacks, and boards—were processed using aqua regia acidic leaching to enhance Cu recovery. Initial results indicated that a concentration of 1% (w/v) from the computer board yielded the highest recovery compared to the capacitor and USB jack parts. The efficiency of Cu leaching was increased by optimizing the duration of the acidic process (from 20 to 60 minutes of leaching time) and the particle size of the e-waste sample (10 mm and 50 mm sizes). The optimum recovery achieved in this study was 2207 mg/L after 20 minutes of leaching time, using a particle size of 10 mm, at 100°C, 150 rpm, and a pulp density of 1% (w/v). Finally, the Cu extracted into the aqua regia solution was recovered at room temperature, 1V, and 0.1-0.5A using the electrowinning process, achieving only 31.6% purity within an hour of electrolysis (difference of 761.6 mg/L of Cu). The lower recovery performance observed in this study could be influenced by the overpotential and strongly acidic pH of the aqua regia solution with Cu extracted. Based on these findings, further investigation and improvement of the factors and parameters influencing metal leaching, as well as the development of safe and environmentally friendly recovery methods, should be pursued in the future.

The authors acknowledge the financial support provided by the Universiti Teknologi MARA (600-RMC/GPM LPHD 5/3 (163/2021)).

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