



Effect of Mechanical Activation in Subsequent leaching of Copper from Waste Printed Circuit Boards; Mini-Review

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ABSTRACT

The aim of this work is to determine the influence of mechanical activation in subsequent leaching of copper from waste printed circuit boards. In this mini -review, the effect of mechanical activation of subsequently copper leaching is discussed in detail based on past research efforts to provide a comprehensive image of the current status. The subsequent leaching of copper species from WPCB powder is considerably improved by mechanochemical activation. To improve copper leaching efficiency from WPCBs, mechanochemical treatment process is suggested.

Keywords:

Mechanical activation; leaching; copper; waste printed circuit boards.

1. Introduction

Since the mid-1990s, electronic waste (E-waste) has been identified as the fastest-growing component of the solid-waste stream worldwide [1]. and a rising number of electronic devices as well as PCs [2]. Furthermore, it is projected that waste printed circuit boards (WPCBs) will make up about 4% of these e-wastes [3, 4].

According to data, the global generation of e-waste in 2016 was over 47 million metric tons, or nearly 4730 Eiffel towers [5]. Similarly, according to the Global E-waste Monitor 2020, the world generated a staggering 53.6 Mt of e-waste as shown in figure 1. Since 2014, worldwide e-waste creation has increased by 9.2 Mt and is expected to increase to 74.7 Mt by 2030, nearly doubling in only 16 years. Higher EEE consumption rates, short life cycles, and limited repair choices are all contributing to the growing volume of e-waste [6].

WPCBs is composed of 30% polymers, 40% metals, and 30% inert oxides in most electronic equipments [4]. WPCBs contain different metals, with copper having the highest percentage (20%) [7].

Based on a comparison of world mine production and reserve base, it was anticipated that copper in minerals will be depleted in 61 years [8]. Copper concentration in WPCBs is significantly more than the 0.2-0.7% in copper ore, making it the primary economic driver for WPCB recycling [9, 10]. More crucially, approximately 4 million tons of copper are recycled for end-of-use goods and manufacturing wastes, with around 22 million tons of copper coming into use each year [10].

Copper in WPCBs is almost always in its elemental form (Cu⁰) [11]. For the recycling of copper from WPCBs, various pyrometallurgical, hydrometallurgical [12], bio-hydrometallurgical [13], physical-mechanical

[14], and electrochemical [15] techniques have been proposed.

Because of the advantages of cheap recovery cost, high metal recovery rate, and moderate operating conditions, hydrometallurgical processes have recently received more attention than pyrometallurgical methods. Metals are dissolved in an alkaline or acid media in hydrometallurgical operations.

Several researchers have investigated the leaching of base metals from WPCBs using sulfuric acid and hydrogen peroxide as oxygen

sources. Apart from this system, nitric acid and hydrochloric acid have been extensively researched, but due to strict environmental restrictions and the caustic nature of these reagents, they cannot be deemed appropriate, whereas sulfuric acid is less harmful [16].

Even when utilizing a mineral acid as the leaching reagent, direct leaching and separation of Cu^0 from WPCB powder is highly difficult [17].



Fig. 1. Global quantity of generated e-waste [6].

Mechanochemical activation applies mechanical energy to condensed matter to induce chemical reactions, and it can also trigger physicochemical changes in solid materials, to improve their leaching activity [18]. Furthermore, because mechanical energy rather than thermal energy drives mechanochemical processing, the extraction of valuable metals from electronic wastes can be completed without harsh conditions such as high temperature or high pressure, achieving the goal of green chemistry for e-waste recycling [19]. Previous investigations have found that pretreatment with mechanical activation improves leaching [20].

2. Printed circuit boards (PCBs)

PCBs mechanically support and link electronic components (ECs) for electrical functions as the most important component within electric and electronic equipment. WPCB recycling is difficult due to its inherent characteristics and complex compositions, which differ significantly from metal recovery from natural minerals [21].

2.1 Chemical compositions of WPCBs

WPCBs are a valuable resource that includes wide a range of metals and are important since their metal concentration is substantially higher than that of ores. WPCBs are widely known for their very diverse makeup. In order to design cost-effective and environmentally friendly recycling systems, the content of valuable materials and dangerous compounds in WPCBs is critical. Nearly 28% of a PCB is metallic and 72% nonmetallic, including ceramics and polymers. There are almost 60 elements in WPCBs. However, the composition of WPCBs varies depending on the origin, age, manufacturer, and class [22].

2.2 Types of PCBs

PCBs are classified as single-sided, double-sided, multi-layer, rigid, flexible, or flex-rigid based on their construction, alignment, and board shape. The structures of PCBs from various EEEs are constantly varied, making WPCB recycling exceedingly difficult. In order to design cost-effective and environmentally

friendly recycling solutions, it is necessary to summarize the many kinds and structures of PCBs. PCBs have a Cu-based fundamental structure. Six substrate fiberglass layers, two isolating fiberglass layers, electrical tracks, and solder masks are all included on the PCBs. The substrate and PCB compositions are classed as FR-4 and FR-2.

3. Pretreatment of WPCBs

Pretreatment is required to separate layers and identify ECs based on their chemical compositions. Pretreatment, complete/selective leaching of metals, purification of metals from leaching solution, and recovery of metals as end products are the steps in the hydrometallurgical recycling of WPCBs.

3.1 Mechanical-physical method

These approaches are thought to be the most ecologically benign for metal recovery [23]. However, because of poor metal separation and low recycling rates, it is generally used for pre-treatment and metal enrichment. Segmentation, size reduction, and separation of the WPBs are the three key steps of mechanical-physical pre-treatment. It should be noted that the pre-treated materials from each stage can be further processed for metal recovery using a hydrometallurgical method [24].

3.2 Chemical Methods

These approaches pertain to the chemical pre-treatment of WPCBs, which is based on the dissolving of several WPCB components to facilitate the downstream leaching process. Because metals are coated by chemical compounds, full separation of metal and non-metal fractions is difficult during mechanical operations. Proper chemical treatments to eliminate the chemical compounds must be investigated. WPCBs are further delaminated utilizing commercially available and low-cost chemical reagents, organic solvents, or ionic solutions to dissolve HERS/BER and chemical coatings [25].

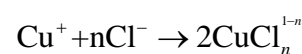
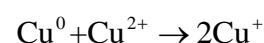
4. Copper leaching from WPCB

Leaching is a necessary step in the hydrometallurgical process to transport copper metal from solid materials to a solution for subsequent recovery.

4.1 Mineral acid leaching of copper

Mineral acids and ammonia-ammonium are two popular leaching techniques. Mineral acids, such as H_2SO_4 , HCl , and HNO_3 , have been frequently reported for metals leaching from WPCBs due to their inexpensive prices, well-studied leaching mechanism, and more flexibility in process control and scalability [26]. Cu can only be leached using oxidizing or non-oxidizing acids containing oxidants (air/ O_2 , H_2O_2 , Cl_2 , Cu^{2+} , Fe^{3+} , and so on), which is explained by their typical reduction potentials. During leaching procedures, the acid concentration is the most important metric [28]. According to Bas and his colleagues, Cu leaching rates increase as HNO_3 concentrations (1–5 M) grow, and a concentration of $\geq 2-3$ M is necessary for a high Cu leaching rate (88.5–99.9%) at 6% pulp density and 70 °C [27].

According to [28], H_2SO_4 and H_2O_2 are the most often used mineral acid and oxidant combinations for full Cu leaching. Yazici and his colleague revealed Cu^{2+} as an oxidant acting with Cl^- in a sulfate solution demonstrating good leaching of Cu (> 90%) as described by Eqs. (1) and (2) [29].



However, the amount of Cl^- should be kept high enough to prevent Cu^+ from precipitating into $CuCl$ ($K_{sp} = 1.86 \cdot 10^{-7}$) (Eqs. (3) and (4)).

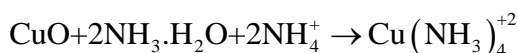
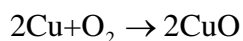


Multi stage leaching has been found to be an effective method for selective metal leaching. Somasundaram et al. extracted Cu from WPCBs using 0.1 M $CuCl_2$ and 3.0 M HCl at 25 °C, with only 6.9% Cu leaching. Cu may then

be leached out in the second step by increasing CuCl_2 concentration and temperature to 0.5 M and roughly 50 °C, respectively [30].

4.1 Ammonia-ammonium leaching of copper metal

A number of investigations have been done using ammonia and ammonium to leach copper metal from WPCBs [31-33]. Cu selectivity and leaching rate ($\geq 90\%$) have been observed for ammonia-ammonium [32, 33]. According to [33], Cu dissolving in ammonia ammonium solution is divided into 2 stages: (i) Cu is initially oxidized at its surface by an oxidant such as dissolved oxygen or H_2O_2 (Eq. (5)); (ii) The ammonia-ammonium solution subsequently dissolves the CuO , resulting in the production of a soluble Cu (I)-ammine complex ($\text{Cu}(\text{NH}_3)_4^{2+}$), as shown in Eq (6).



The author of [34] employed an 8% ammonia-0.5 M ammonium citrate solution with a liquid-to-solid ratio of 10:1 to leach over 98% Cu.

1.5 Effect of mechanochemical treatment on Cu^0 leaching

For the first time, Zhiyuan Ou and Jinhui Li looked into using mechanochemistry to recover copper from waste printed circuit boards (WPCBs) [35]. Their goal was to recycle as much copper (9.89 wt%) and resin (the two primary components of WPCBs) as possible. In the model experiment, copper and sulfur could be interacted to generate copper sulfides via mechanochemistry, so the synergistic method of mechanical ion activation and sulfurization was proposed. The yield of copper reached about 90% after milling WPCB fragments and sulfur for 20 minutes and leaching the milled sample in sulfuric acid (3 M) and hydrogen peroxide (30 wt%).

Weihua Gu et al. advocated using mechanical activation to improve *Acidithiobacillus ferrooxidans* bioleaching efficiency and to extract metals from WPCBs

prior to bioleaching [7]. The best conditions for mechanical activation were a milling time of 2 hours, a milling speed of 340 rpm, and a ball to powder ratio was 10:1; the Cu bioleaching rate was 94.33 %. Without mechanical activation pretreatment, the Cu bioleaching rate was 74.75%. Mechanical activation, they discovered, might lead to lower particle sizes and reveal wrapped metals, enhancing the bioleaching effectiveness of the metals inside the WPCBs. Mechanical activation was observed to increase the rate of Cu bioleaching from WPCBs, indicating that combining mechanical activation with a biological technique to extract metals from WPCBs is a potential technology.

Kang Liu and his colleagues studied the influence of mechanochemical activation factors on copper recovery from WPCBs [11]. In the cupric sulfate regeneration process, $\text{K}_2\text{S}_2\text{O}_8$ was used as the only reagent instead of acid/alkali reagents. They investigated the impacts of several mechanochemical processing factors on Cu recovery and $\text{K}_2\text{S}_2\text{O}_8$ breakdown %, including time, $\text{K}_2\text{S}_2\text{O}_8$ /WPCBs mass ratio, ball-to-powder ratio, and ball-milling speed. The leaching of Cu species from WPCB powder was shown to be greatly aided by mechanochemical processing. Cu recovery percentage was reached 90.1 wt% at the end of 4 hours.

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