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# Copper leaching from waste printed circuit boards using typical acidic ionic liquids recovery of e-wastes' surplus value

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ABSTRACT

In this study, using several types of acidic ionic liquids as the leaching reagents, the leaching behaviors of the copper present in waste printed circuit boards (WPCBs) were investigated. The effects of various parameters on the copper leaching rate were studied, such as the particle size of the shredded WPCB, type of ionic liquid used, hydrogen peroxide dosage, solid-to-liquid ratio, leaching temperature, and leaching time. The experimental results showed that the copper leaching rate increases continuously when the powder particle size is increased from 0.071 to 0.500 mm. Moreover, the copper leaching rate also increases with an increase in the leaching temperature. In contrast, the leaching rate first increases and then decreases with increases in the leaching time, hydrogen peroxide dosage, and solid-to-liquid ratio. The optimal conditions that provided a 98.31% copper leaching rate were: particle size >0.500 mm, 8.5 mL 90% (v/v) ionic liquid, 1.5 mL 30% hydrogen peroxide, solid-to-liquid ratio of 1/20, leaching temperature of 80 °C, and leaching time of 2 h.



In recent years, with the advancement of technological innovation and the acceleration of the replacement of electronic appliances, more and more electronic waste has been created. The world's output in 2017 was 40 million tons; this not only harms the environment but also causes human health issues. Thus, solving the issues of electronic waste, especially the recycling of waste CPUs, has become an urgent environmental and economic requirement.

With regard to the process of recovering metals from utilized circuit boards, we specifically studied the recovery of metallic copper, and compared the use of ionic liquids, mechanical methods, fire, biological methods, and supercritical fluid methods in terms of yield and recovery efficiency, differences in environmental protection, toxicity, and reaction conditions. Experimental results show that the fastest copper recovery time from waste circuit boards was 2.5 h by using ionic liquids. Furthermore, dioxins are produced by the fire method, the biological method requires of the use of toxic bacteria, and the supercritical fluid method has very strict requirements on temperature and pressure. In comparison, the ionic liquid reaction conditions are relatively mild and easy to implement.

Ionic liquids have several useful properties: (1) wide liquid range, from below or near room temperature to above 300 °C, with high thermal and chemical stability; (2) low vapor pressure and no volatilization means that evaporation does not occur during storage and use; (3) can be reused many times; (4) eliminates the production of toxic volatile organic compounds (VOCs); (5) High conductivity and large electrochemical window mean that they can be used as electrolytes for the study of many substances; (6) the design of anions and cations can be used to adjust its solubility to inorganic substances, water, organics, and polymers; and (7) a large degree of polarity controllability, low viscosity, high density, and the ability to form two or more systems, means they are suitable for separation of solvents or constitute a new system of reaction-separation coupling.

In this study, the leaching rate was 94.30% when the solid-liquid ratio was 1/20, leaching time was 2 h, and temperature was 80 °C. The ionic liquid consumption was relatively lower than pyrometallurgical separation, the time was shorter, the efficiency was higher, and involved lower energy consumption. The study is of great significance for industrial production.

## 1. Introduction

Printed circuit boards can cause serious environmental damage after disposal (Rüşen and Topçu, 2017). Waste Printed Circuit

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Boards (WPCBs) present a continuous growth trend with an increase rate of 17–25% per year, and a total of 65.4 million tons of waste electric and electronic equipment (WEEE) are generated annually worldwide (Tatariants et al., 2017). WPCBs account for about 3% by weight of the total amount of electronic waste, bring a global challenge for the environment because of their complex and hazardous components (Wang et al., 2018). WPCBs contain large amounts of metal fractions, such as Cu 20%, Fe 8%, Ni 2%, Sn 4%, Pb 1%, Al 2%, Zn 1%, Sb 0.4%, Au 500 g/t, Ag 1000 g/t, and Pd 50 g/t, and include epoxy resin, glass fibers, ceramics and other nonmetallic fractions which embody large numbers of brominated flame retardants and other harmful substances (Li et al., 2014; Liu et al., 2017; Zhou et al., 2016). Therefore, the recycling of WPCBs is an immediate problem due to serious threats to human health and the surrounding environment (Debnath et al., 2016).

The high economic value of metal has undoubtedly contributed in attracting more attraction to this issue. Thus, many recycling methods have been successively utilized to reclaim metals from WPCBs, such as mechanical separation, pyrometallurgical separation, bioleaching separation (Zhu et al., 2011), supercritical fluid separation, and hydrometallurgical separation (Leite et al., 2017). Mechanical separation has low efficiency in recovering precious metals because of the purification step required after obtaining the metal mixtures (Cui and Zhang, 2008; Huang et al., 2014); as a result, mechanical separation is usually used as the pretreatment process. Pyrometallurgical separation became the main method used for a long time, but was eventually eliminated due to toxic gases, dioxins, furans, and other poisonous substances produced during combustion (Birloaga and Vegliò, 2016). The third common method is bioleaching separation, which is a promising alternative to extract metals from WPCBs (Davris et al., 2018). However, the presence of nonmetallic fractions encouraged bacterial toxicity during the leaching process, which reduced the leaching rate (Chen et al., 2015b; Ilyas et al., 2010). Furthermore, longer leaching time and specific bacterial species that were harder to obtain prevented the promotion of this method. Another method uses supercritical fluids with negligible low surface tension, high diffusivity, low viscosity, and ability to dissolve inorganic salts (Liu et al., 2016). The disadvantage is that the method requires higher critical temperature and pressure which may result in significant reactor corrosion and higher energy consumption (Yousef et al., 2018). It should be noted that the use of ionic liquids to leach copper from waste circuit boards is more environmentally friendly than pyrometal recovery of copper. Furthermore, it is more efficient than mechanical recovery of copper metal that achieves 79.54% yield of the heavy liquid separation process to recover copper, which is generally lower than the 90–99.84% yield acquired via ionic liquid. Additionally, its toxicity is lower than the biological method, which typically recovers only 76.59%. At the same time, its reaction conditions are milder than the supercritical method and the required reaction time at 2 h is shorter than the supercritical method that requires 54 h.

Room temperature ionic liquids (RTILs) are essentially in the liquid state at low temperatures (Kim et al., 2018), and typically contain organic cations and inorganic or organic anions. Ionic liquids have a wide liquid temperature range and many other unique properties such as negligible volatility, low vapor pressure, high thermal stability (<400 °C), high electrical conductivity, and wide electrochemical windows (Zhu et al., 2016). As a result, ionic liquids are increasingly employed in fields including biochemical process, organic synthesis processes (Zhu et al., 2012), separation process, catalytic reaction, and materials engineering (Ma et al., 2012). Previous study showed that ionic liquids can usually be reused more than 10 times (Zhang et al., 2013). Thus, ionic liquids are very promising as an environmentally friendly reagent for leaching metals from WPCBs (Dong et al., 2009).

The first step in dismantling WPCBs is to remove the solder. Commonly, the IL 1-butyl-3-methylimidazolium tetrafluoroborate ([BMIM]$BF_4$) (Bar-Cohen et al., 2017) was used as the heating medium to recycle the valuable solder material from WPCBs (Zeng et al., 2013). Furthermore, 1-carboxymethyl-3-methylimidazolium bisulfate ([CM-MIM]$HSO_4$) can be used as an alternative IL for recycling metals from WPCBs under different conditions, such as different leaching times, leaching temperatures, solid–liquid ratios, WPCB powder particle sizes (Zhang et al., 2018), and hydrogen peroxide dosages.

## 2. Materials and experimental methods

### 2.1. Preparation of WPCBs and removal of solder

The WPCBs with electronic components mounted on them were pretreated as shown in Fig. 1. They were first cut into small pieces with dimensions of approximately 50 × 50 mm using a cutting machine. Then, these pieces were shredded using a cutting mill and sieved into different fractions using standard sieves: $F_1 < 0.071$ mm, $0.071 < F_2 < 0.100$ mm, $0.100 < F_3 < 0.250$ mm, $0.250 < F_4 < 0.500$ mm, and $F_5 > 0.500$ mm. Next, the sieved parts were dried at 105 °C for 24 h.

As is shown in Fig. 2, the process of dismantling the WPCBs was as follows. The IL [BMIM]$BF_4$ was heated to 200 °C. Then, while keeping the temperature uniform, a certain mass of the cut WPCB pieces was added to the IL, which was stirred with a mechanical stirrer at a rate of 150 rpm until all the solder points in the WPCBs had melted. As a result, owing to gravity and the centrifugal force, the molten solder dropped from the circuit board, resulting in the separation of the circuit board substrate and the electronic components. The main parameters for the recovery of the solder from the WPCBs were temperature of 200 °C, mechanical stirring speed of 150 rpm, and melting time of 3 min.

### 2.2. Leaching experiments

After recycling the solder, the next step was crushing the WPCBs into powders, followed by leaching the metals using ILs.

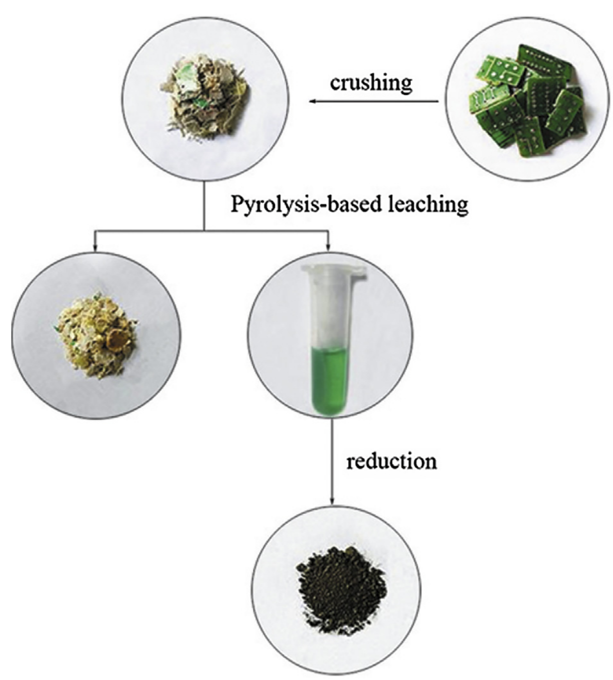


**Fig. 1.** Schematic of process for recovering metals from WPCBs using ILs.