Release kinetics of Ba and Pb contained in WPCBs during the traditional hydrometallurgical process

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Abstract

The release kinetics of heavy metals (Ba and Pb) contained in WPCBs, during the traditional hydrometallurgical process, were investigated in this study. The effects of different conditions, i.e. particle size, solid/liquid ratio, amount of H\textsubscript{2}O\textsubscript{2} and HNO\textsubscript{3}, HNO\textsubscript{3} concentration, react time and temperature, on Ba and Pb release were examined. The highest release rates of Ba and Pb were obtained. For Ba, particle size was <100 mesh, solid/liquid ratio was 1:2, HNO\textsubscript{3} concentration (HNO\textsubscript{3}:H\textsubscript{2}O\textsubscript{2}, v/v) was 1:2, amount of both H\textsubscript{2}O\textsubscript{2} and HNO\textsubscript{3} were 6 mL, react time was 4 h and temperature was 50 °C. For Pb, particle size was 40-20 mesh, solid/liquid ratio was 1:3, HNO\textsubscript{3} concentration was 1:3, amount of both H\textsubscript{2}O\textsubscript{2} and HNO\textsubscript{3} were 4 mL, react time was 2 h and temperature was 50 °C. The results showed that the highest release concentration of Ba and Pb were up to 76.89 g/L and 2.18 g/L, respectively.

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Keywords: WPCBs; hydrometallurgical; release; concentration; heavy metals; Ba; Pb

1. Introduction

Printed circuit boards (PCBs) are widely used in almost all the electric and electronic equipments (EEE) as an essential part, such as P.C., household appliances, etc. With rapid economic growth in the past few years, PCBs manufacturing has been developed dramatically. It is estimated that the average rate of PCBs manufacture increased by 8.7% in the world, and the number is much higher in China (14.4%)\textsuperscript{[1]}, which results in a huge quantity of waste printed circuit boards (WPCBs). WPCBs has been attracting the
public attention by its abundant valuable matters (i.e., plastic, resins and metals etc.) as well as environmentally harmful substance (i.e., heavy metals and halogenated flame retardants, etc.). Especially, the heavy metals contained in WPCBs, such as zinc, nickel, barium and lead, may pollute the environment and affect human health seriously if they are not properly disposed [2]. Therefore, recycling of WPCBs has become an important issue, not only with regard to the recovery of valuable materials, but also from the perspective of the environmentally harmful [3].

In recent years, many researchers have investigated a number of approaches on copper recovery from WPCBs, including mechanical, pyrometallurgical, bioleaching and hydrometallurgical processes [4-9]. So far, the hydrometallurgical processes have been widely applied to recover metals from WPCBs due to the high metal recovery [5]. Leaching is the first step in recovery of metals from WPCBs using hydrometallurgical method [8], and plenty of related studies were mainly focused on the recovery of main metals such as copper, gold and lead [10-12]. However, few studies were reported on investigating the release of heavy metals by leaching with hydrometallurgical process.

In this regard, the release of heavy metals (barium and lead) in the leaching process was investigated in the present paper. And the effects of different conditions on the release of heavy metals were discussed as well.

1. Materials and experimental

2.1 Materials

WPCBs used in this study were obtained from waste computers. After the electric and electronic components (relays, capacitors, etc.) were disassembled, the WPCBs were sent to comminute in a cutting mill until the fractions reached particle size smaller than 0.5 mm. The WPCBs powder obtained was dried at 105°C for 24 h.

Nitric acid, hydrogen peroxide (30 wt.%) and hydrofluoric acid were used to fully leach the metals from the WPCBs specimen by microwave digestion [13]. The metal concentrations of the digestion solution were tested by Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES, Thermo Scientific, iCAP 6500). The results are presented in Table 1. It can be seen from Table 1 that the content of barium and lead were 3.67% and 1.80%, respectively.

| Element | Cu | Ca  | Al  | Ba  | Pb   | Sn  |
|---------|----|-----|-----|-----|------|-----|
| Content (wt.%) | 22.82 | 11.84 | 7.00 | 3.67 | 1.80 | 1.28 |

1.2 Experiments
All experiments were carried out in a 250 mL glass conical flask placed in a constant temperature water bath oscillator, using a constant oscillating frequency of 250 rpm in the temperature range of 40 to 90 °C. Concentration of heavy metals (barium and lead) in the leach liquor was analyzed by ICP-OES. The SEM (Scanning Electron Microscope, Leica Cambridge LTD, S440) images of WPCBs and leach residue are presented in Fig. 1.

![SEM images of (a) WPCBs samples (Particle size of WPCBs was 40-100 mesh.) and (b) leaching residue (Both the amount of H₂O₂ and HNO₃ were 2 mL, the HNO₃ concentration, solid/liquid ratio (WPCBs/ H₂O, g/mL), leaching time and temperature were 1:3 (HNO₃:H₂O, v/v), 1:2, 2 h and 50 °C respectively.)](image)

Fig. 1. SEM images of (a) WPCBs samples (Particle size of WPCBs was 40-100 mesh.) and (b) leaching residue (Both the amount of H₂O₂ and HNO₃ were 2 mL, the HNO₃ concentration, solid/liquid ratio (WPCBs/ H₂O, g/mL), leaching time and temperature were 1:3 (HNO₃:H₂O, v/v), 1:2, 2 h and 50 °C respectively.)

2. Results and discussion

As is seen from Fig. 2, decreasing the particle size from >20 mesh to <100 mesh increased the Ba release concentration from 18.49 g/L to 76.89 g/L. However, the release concentration of Pb was increased from 0.47 g/L to 1.11 g/L when the particle size decreased from >20 mesh to 40-100 mesh, and decreased when the particle size was less than 100 mesh.

3.1 Effect of particle size

2 g WPCBs were used in this series of experiments. Amount of both HNO₃ and H₂O₂ were 4 mL, HNO₃ concentration (HNO₃:H₂O, v/v), solid/liquid ratio (WPCBs/ H₂O, g/mL), reacting time and temperature were 1:3, 1:3, 2 h and 50 °C respectively. While the investigated particle size were >20 mesh, 40-20 mesh, 100-40 mesh and <100 mesh. The effect of particle on release of Ba and Pb were given in Fig. 2.
2.2 Effect of solid/liquid ratio

The WPCBs with particle size of 40-20 mesh was used and the effect of solid/liquid ratio on release of Ba and Pb were shown in Fig.3. Other conditions remained constant and the examined solid/liquid ratios were 1:1, 1:2, 1:3, 1:5 and 1:7.

It can be found from Fig.3 that the release concentration of Ba and Pb showed the same tendency. The release concentration of Ba and Pb reached the highest points when the solid/liquid ratio was 1:2, and the release concentration of Ba and Pb were 26.58 g/L and 2.18 g/L, respectively.
2.3 Effect of addition of hydrogen peroxide

When solid/liquid ratio remained 1:3, other conditions were kept the same as previously, the effect of H$_2$O$_2$ amount on release of Ba and Pb was given in Fig. 4. The investigated H$_2$O$_2$ amounts were 2 mL, 4 mL, 6 mL and 8 mL.

![Fig. 4. Effect H$_2$O$_2$ addition on release of Ba and Pb.](image)

From Fig. 4, it can be seen that the effect of H$_2$O$_2$ amount on release concentration of Ba and Pb showed the same tendency, just as the effect of solid/liquid ratio. When addition of H$_2$O$_2$ was 6 mL, the highest release concentration of Ba and Pb were 25.48 g/L and 1.61 g/L, respectively.

2.4 Effect of HNO$_3$ amount

Addition of H$_2$O$_2$ was 4 mL, other conditions were kept the same as previously, the effect of HNO$_3$ amount on release of Ba and Pb was given in Fig. 5. The investigated HNO$_3$ amounts were 2 mL, 4 mL, 6 mL and 8 mL.
Fig. 5. Effect of HNO$_3$ amount on release of Ba and Pb.

It can be seen from Fig. 5 that the release concentration of Ba and Pb reached highest when HNO$_3$ amount was 6 mL, and the highest release concentration of Ba and Pb were 41.18 g/L and 1.36 g/L, respectively.

2.5 Effect of HNO$_3$ concentration

HNO$_3$ amount was 4 mL and other conditions were kept the same as previously, the effect of HNO$_3$ concentration (HNO$_3$:H$_2$O, v/v) on release of Ba and Pb was given in Fig. 6. The investigated HNO$_3$ concentrations were 1:2, 1:3, 1:4, 1:5 and 1:6.
As is shown in Fig. 6, decreasing the HNO₃ concentration from 1:2 to 1:6 decreased the Ba release concentration from 24.43 g/L to 15.07 g/L. However, the release concentration of Pb was reached highest when HNO₃ concentration was 1:4, and the release concentration was 1.88 g/L.

2.6 Effect of react time

Other conditions were kept the same as previously, and HNO₃ concentration remained 1:3, the effect of react time on release of Ba and Pb was given in Fig. 7.
It can be found from Fig. 7 that the react time did not affect the release concentration of Ba significantly. However, increasing the react time from 2 h to 10 h decreased the Pb release concentration from 1.05 g/L to 0.10 g/L. And the highest release concentration of Ba and Pb were 23.75 g/L and 1.05 g/L, when the corresponding react time was 4 h and 2 h, respectively.

2.7 Effect of temperature

When react time was set at 2 h and other conditions were kept the same as previously, the effect of temperature on release of Ba and Pb was given in Fig. 8.
From Fig. 8, it can be seen that increasing the temperature from 20 °C to 50 °C increased the release concentration of Ba from 0.71 g/L to 22.52 g/L and reached highest. And the release concentration of Pb was reached highest when the temperature was 30 °C, and the release concentration was 1.14 g/L.

4. Conclusion

(1) The heavy metals (Ba and Pb) in WPCBs were released in hydrometallurgical process, and the release concentration of Ba and Pb were affected significantly in the present react conditions.

(2) Particle size, solid/liquid ratio, amount of H$_2$O$_2$ and HNO$_3$, HNO$_3$ concentration, react time and temperature were investigated in the present study. The highest release concentration of Ba were obtained when particle size was <100 mesh, solid/liquid ratio was 1:2, HNO$_3$ concentration (HNO$_3$:H$_2$O, v/v) was 1:2, amount of both H$_2$O$_2$ and HNO$_3$ were 6 mL, react time was 4 h and temperature was 50 °C. For Pb, particle size was 40-20 mesh, solid/liquid ratio was 1:3, HNO$_3$ concentration was 1:3, amount of both H$_2$O$_2$ and HNO$_3$ were 4 mL, react time was 2 h and temperature was 50 °C.

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