ARTICLE IN PRESS

Journal of Cleaner Production xxx (2015) 1–6

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Lead during the leaching process of copper from waste printed circuit boards by five typical ionic liquid acids

Mengjun Chen*, Sha Zhang, Jinxiu Huang, Haiyan Chen

Key Laboratory of Solid Waste Treatment and Resource Recycle (SWUST), Ministry of Education, Southwest University of Science and Technology, 59 Qinglong Road, Mianyang, 621010, China

ARTICLE INFO

Article history:
Received 2 December 2014
Received in revised form
1 February 2015
Accepted 17 February 2015
Available online xxx

Keywords:
WPCBs
Ionic liquids (ILs)
Lead
Leaching kinetics

ABSTRACT

Waste printed circuit boards (WPCBs) contain both valuable resources and toxic substances, causing serious pollution to the environment if not treated appropriately. However, currently almost all researches are focused on valuable resources recovery, mainly copper, without considering the effect of toxic substances, such as lead, during the recycling process. Hence, we selected lead to represent the heavy metals and examined its behavior in typical ionic liquid (IL) acids leaching system. The factors that affect lead leaching rate, such as particle size, temperature, ionic liquid concentration, H_2O_2 adding amount and solid to liquid ratio, were examined in detail. The results show that particle size, IL acid concentration, solid to liquid ratio and hydrogen peroxide dosage hardly affect lead leaching rate for $[BSO_4HPy]HSO_4$, $[BSO_3HMIm]HSO_4$ and $[MIm]HSO_4$, while lead leaching rate is significantly influenced by these factors for $[BSO_3HMIm]OTf$ and $[BSO_3HPy]OTf$. For copper, it is totally on the opposite. It is hard to discuss the leaching kinetics for lead, while for the leaching of copper, diffusion plays a more important role than surface reaction. Most importantly, compared to lead, IL acids seem to show a selectivity for copper leaching out from WPCBs. Therefore, IL acids could be a prospective choice for leaching copper from WPCBs.

1. Introduction

Printed circuit board (PCB) is an important part of electronic industry. From personal computers, TVs, communication equipment to electronic toys, almost all of the electronic products contain circuit board. Both technological innovation and market expansion accelerate the replacement of electric and electronic equipment (EEE), which leads to a significant increase of waste PCBs (WPCBs). WPCBs have been attracting the public attention by its environmentally harmful materials and abundant valuable nonferrous metals. It contains plenty of toxic substances, such as brominated flame retardants (BFR), PVC plastic and heavy metals. In general, WPCBs contain approximately 30% metals and 70% nonmetals. Especially, the typical metals in WPCBs consist of copper (20%), iron (8%), tin (4%), nickel (2%), lead (2%), zinc (1%), silver (0.2%), gold (0.1%), and palladium (0.005%) (Huang et al., 2009). The purity of precious metals in WPCBs is at least 10 times higher than that of rich-content minerals (Guo et al., 2008a). Thus, recycling of waste PCBs has become an important issue in the world, not only from the perspective of environmentally harmful, but with regard to the recovery of abundant valuable materials (Quan et al., 2010).

Currently, techniques such as chemical, biological and mechanical methods are available for recycling WPCBs. Mechanical methods, including crushing, vibration and electrostatic separation, are extensively applied to separate the metals from the nonmetals of WPCBs (Guo et al., 2008b; Wu et al., 2008; Duan et al., 2009). Therefore, the mechanical process is usually used as pretreatment. Some researchers have studied metal recovery from WPCBs with the aid of chemical and biological processes, such as hydrometallurgy, pyrometallurgy, electrolysis and bioleaching (Veit et al., 2006; Moltó et al., 2009; Yang et al., 2011). For the bioleaching process, the presence of non-metallic components affected the recovery rate of metals, thus how to improve metals recovery in bioleaching from WPCBs need further research. Leaching, as the first step in the extraction of metals by hydrometallurgical process, has been given more attention, including nitric acid (Hoang et al., 2011), ammoniacal sulfate (Guo et al., 2008b) and chloride (Yang et al., 2011).

Room temperature ionic liquids (RTILs), also called ionic liquids and considered as the green solution for the future, which are

* Corresponding author. Tel./fax: +86 816 2419569.
E-mail address: kyling@swust.edu.cn (M. Chen).

http://dx.doi.org/10.1016/j.jclepro.2015.02.045
0959-6526/

Please cite this article in press as: Chen, M., et al., Lead during the leaching process of copper from waste printed circuit boards by five typical ionic liquid acids, Journal of Cleaner Production (2015), http://dx.doi.org/10.1016/j.jclepro.2015.02.045

basically liquid at low temperature typically consisting of an organic cation with an inorganic or organic anion and have a wide liquids temperature range (Dong et al., 2009). Currently, ionic liquids are mainly used in the solid waste field of biomass resources. In 2012, zhu et al. for the first time used $[EMIM]^+[BF_4]^-$ to dissolve bisphenol A diglycidyl ether contained in WPCBs and achieved the separation and recovery of copper foil, glass fibers and solder at the same time (Zhu and Chen, 2012a, 2012b, 2012c). Meanwhile, the ionic liquid $[EMIM]^+[BF_4]^-$ can be regenerated by washing (Zhu and Chen, 2012a, 2012b, 2012c), which opened a new stage in the disposal of electronic waste by ionic liquids. After that Huang et al. reported that copper can be successfully leached out from WPCBs by an ionic liquid acid, 1-butyl-3-methyl-imidazolium hydrogen sulfate ([bmim]HSO_4) (Huang et al., 2014). However, all these researches were focused on the recovery of valuable resources, mainly copper, few work has been reported on the behavior of hazardous metals, such as lead, during the recycling process.

Based on the work of Huang et al., we further examined other five typical IL acids, [BSO_4HPy]HSO_4, [BSO_3HMIm]HSO_4, [BSO_3HMIm]OTf, [BSO_3HMIm]OTf and [BSO_3HPy]OTf, to see if these IL acids could successfully leach copper out from WPCBs. In order to investigate the behavior of lead during the leaching process by IL acids, a study is conducted to test if lead could be leached out by these IL acids simultaneously as copper and to discuss the effect of lead on copper leaching. Factors that affect the leaching efficiency of lead, such as granularity of WPCBs, ionic liquid acid concentration, solid–liquid ratio, leaching time and temperature, have been studied in detail. At the same time, the data for copper, provided by supporting information (SI), was introduced to examine the relationship between lead and copper.

2. Materials and methods

2.1. Sample preparation

First, WPCBs were cut into small pieces, around 50 mm × 50 mm. Then it was further shredded using the Retsch SM-2000 Cutting Mill (Retsch, Germany), and sieved into different fractions using standard sieves: F1 < 0.075 mm, 0.075 mm < F2 < 0.1 mm, 0.1 mm < F3 < 0.25 mm, 0.25 mm < F4 < 0.5 mm and F5 > 0.5 mm. After that, the obtained powders were digested by microwave aided HNO_3–H_2O_2–HF system (Yamasaki, 1997). Lead concentration of the digested solutions were analyzed by Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES, Thermo Scientific, iCAP 6500) and the results are given in Table 1.

2.2. Leaching experiments

All the leaching experiments were carried out in a batch of 250 mL glass conical flasks placed in a constant temperature water bath oscillator, using a constant oscillating frequency of 250 rpm at 40–70 °C. Five ionic liquids, [BSO_4HPy]HSO_4 (N-sulfobutylpyridinium hydrosulfate), [BSO_3HMIm]HSO_4 (1-sulfobuty-3-methylimidazolium hydrosulfate), [BSO_3HMIm]OTf (N-sulfobutylpyridinium trifluoromethanesulfonate), [MIm]HSO_4 (Methylimidazolium hydrosulfate) and [BSO_3HPy]OTf (1-sulfobutyl-3-methylimidazolium trifluoromethanesulfate), were used in the experiment. Their structures are given in Fig. 1. The experiment oxidant is hydrogen peroxide (30 wt.%). Chemicals used in the experiments were all analytical reagents unless otherwise mentioned. Factors that affect the leaching efficiency for the five IL acids were investigated separately, which were WPCBs particle size, IL acid concentration (V/V), IL acid adding amount, H_2O_2 adding amount, temperature and time, and the detailed experiment arrangements are given in Table 2. Reproducibility was guaranteed by repeating each experimental run at least twice. The relative standard deviations of the triplicates were in the limited ranges of a certified commercial laboratory and mean values are given in the Tables and Figures without error bar.

Lead leaching rate is expressed as the lead extracted from the original sample to the proportion of the leaching solution, which is calculated according to the following formula:

$$\text{lead leaching rate} = \frac{\text{lead extracted into leaching solution}}{\text{total lead in waste PCBs specimens}} \times 100\% \quad (1)$$

3. Results and discussion

3.1. Effect of particle size

Fig. 2 shows the effect of particle size on lead leaching rate. As it can be seen in Fig. 2, the leaching rate of lead increases with the increase of particle size. Take [BSO_3HMIm]OTf for example, lead leaching rate increases from 0.83% to 41.44% when WPCBs particle size increases from <0.075 mm to >0.5 mm. For the rest four IL acids, lead leaching rate is not significantly affected by particle size changes in the whole investigated range. For instance, lead leaching rate for [MIm]HSO_4 slightly increases from 1.15% to 4.53% as particle size increasing from <0.075 mm to >0.5 mm.

Under the same conditions, compared to lead, copper leaching rate is significantly affected by WPCBs particle size (SI, Fig. S1). For example, for [MIm]HSO_4, copper leaching rate increases rapidly from 8.85% to 75.71% when the particle size increases from <0.075 to >0.5 mm, while lead leaching rate is slightly changed in the range of 1.15%–4.53%. Second, copper leaching rate possesses an effective size range and this phenomena is consistent with the results reported by Zhu et al. (Zhu et al., 2011), who found that the maximum leaching efficiency of copper was obtained when the particle size was about 18 mm–25 mm. This is reasonable since the smaller the particle size, the bigger surface area per unit mass and particle–particle collision would greatly increase if the particle size is bellow a critical level.

3.2. Effect of ionic liquid acid concentration

The effect of IL acid concentration on lead leaching rate is shown in Fig. 3. In general, IL acid concentration has a pronounced effect on metal extraction. For different ionic liquid, the effect on lead leaching rate is different. [BSO_3HMIm]OTf and [BSO_3HPy]OTf show greater effect than the other three IL acids. Lead leaching rate by [BSO_3HMIm]OTf is in the range of 6.98%–20.04%, while it is around 3% for [BSO_4HPy]HSO_4, [BSO_3HMIm]HSO_4 and [MIm]HSO_4. The acidity of [BSO_4HPy]HSO_4, [BSO_3HMIm]HSO_4 and [MIm]HSO_4 should be higher than [BSO_3HMIm]OTf and [BSO_3HPy]OTf because

Table 1
Lead concentration in the five fractions of WPCBs by ICPOES.

Particle size, mm	F1 (<0.075 mm)	F2 (0.075–0.1 mm)	F3 (0.1–0.25 mm)	F4 (0.25–0.5 mm)	F5 (>0.5 mm)
Pb wt. %	1.67	0.97	1.29	1.42	0.62

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