



Selective copper recovery from complex mixtures of end-of-life electronic products with ammonia-based solution



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ABSTRACT

Electronic waste treatment within EU has focused on valuable metal recycling and has been defined by the EU directives. The difficulties for recycling are induced by the high complexity of such waste. In this research, a hydrometallurgical process was developed to recycle copper from industrially processed information and communication technology (ICT) waste. By using air as the oxidant and ammonia–ammonium carbonate leaching solution, copper could be extracted with high recovery – more than 90%, and high extraction selectivity – around 98%. In order to understand the copper extraction process and the reaction mechanisms, the effects of a range of parameters during copper leaching were comprehensively investigated, including ammonia concentration, leaching temperature, ammonium carbonate concentration, the liquid-to-solid ratio, air flow rate and mechanical stirring rate. The controlling step for the leaching kinetics was identified and the effects of different parameters were investigated. This research is potentially beneficial for further optimisation of the copper leaching process and the whole process design for copper recycling after incorporating with solvent purification and electrowinning of the copper-rich solution.

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1. Introduction

Electronic waste or waste electrical and electronic equipment (WEEE) is considered to be one of the fastest growing solid wastes with an increase of 3–5% annually in the world (Bigum et al., 2012; Cui and Zhang, 2008; Kiddee et al., 2013). The WEEE directive (2012) in Europe has distinguished the environmental and recovery significances of different WEEE and the minimum recovery targets have been clearly defined. It is further promoting research on WEEE treatment technologies, especially valuable or strategic metal recovery from electronic wastes. Concerning WEEE treatment, numerous researches have been performed and abundant literature including critical reviews is available. A typical industrial process is based on high temperature metallurgy including high temperature burning of organics, smelting of metal and oxides, refining and further electrochemical treatment after chemical leaching (Bigum et al., 2012). Comparing with high-temperature pyrometallurgical processes, a low temperature hydrometallurgical process is always preferable especially for WEEE with low calorific values (Fleming, 1992; Ha et al., 2010; Havlik et al., 2011; Jha et al., 2001; Shuva and Kurny, 2013). During hydrometallurgical processing, both acidic and alkaline lixiviant systems have been used and usually additional separation

steps will be included to recover different metals (Koyama et al., 2006; Tuncuk et al., 2012). However, leaching selectivity and effectiveness are frequently the concerns. In addition, the research is mostly focused on mono-streams or relatively 'clean' WEEE, for instance, printed circuit boards (PCB) (Koyama et al., 2006; Li et al., 2007), batteries (Shuva and Kurny, 2013) and mobile phones after disassembly (Li and Xu, 2011). Processing of a more complex type of electronic waste, usually mixtures of all kinds of end-of-life products or wastes from different streams, however, is still a challenge. This type of electronic waste is recently becoming one of the main WEEE feedstocks. Development of a process of less energy-consumable, less sensitive to waste inputs, highly effective and closed-loop, by integrating fundamental metallurgy principles, is therefore of great importance due to the current status of WEEE management and resource constraints.

In our previous work, the possibility of metal extraction selectively from a highly complex industrial information and communication technology (ICT) waste by using a hydrometallurgical method has been presented (Xiao et al., 2013). Copper in the ICT waste could be effectively extracted by using ammonia–ammonium carbonate/sulphate solution with the addition of hydrogen peroxide (Xiao et al., 2013). It was also found that the energy consumption can be minimised by carefully controlling the proton activity and the oxygen partial pressure where Cu^{2+} can also act as the oxidant (Koyama et al., 2006). In this research we demonstrate the potential of development of a hydrometallurgical process for ICT waste recycling to effectively recover copper by using air as

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the main oxidant at room temperature in an ammonia–ammonium carbonate solution. As the essential basis for building up a near to industry process, this research focuses on the apparent leaching behaviour of copper during the copper extraction step which is the main content of the ICT waste in a closed-loop process. The examined parameters during copper leaching include liquid-to-solid ratio (L_s), reagent concentrations (C), mechanical stirring rate (w), temperature (T) and air flow rate (r). As one of the main parts of ICT waste recovery, copper apparent leaching kinetics provides significances on understanding the important factors, the controlling steps for metal extraction as well as the reaction mechanisms which are essential for further process design.

2. Experimental

2.1. Materials

The ICT waste is provided by Van Gansewinkel Groep (VGG). The material has been pretreated and concentrated physically and shredded into size of smaller than or equal to 8 mm. This size is defined to be a limit since further shredding will significantly increase the processing cost. The material is inherent with high inhomogeneity from observation and contains a large fraction of sand, stones, glass, ceramics and plastic particles. Metallic materials are either trapped in non-metallic components or present as metal wires. The general morphology and compositions are given elsewhere (Xiao et al., 2013). Ammonia (25% in water, Alfa Aesar) and ammonium carbonate (99.0%, Alfa Aesar) are used in the experiments.

2.2. Experimental set-up and procedures

The as-received ICT waste was directly used as the feedstock for metal extraction. In each lab-scale experiment, a sample size of around 50 g was taken which would be a relatively representative sample size of the bulk composition according to our previous investigations (Xiao et al., 2013). The leaching experiments were carried out in a glass reactor as shown in Fig. 1. The round shape of the reactor could help to ensure a better mixing condition. Air was purged into the bottom of the reactor and the gas bubbles could be further broken up by the agitator. The gas flow rate was controlled by a flow metre. During leaching, system temperature is slightly fluctuating because of the exothermic reactions (Xiao et al., 2013). A heating bath/thermo-stat with water was used to control the temperature and an additional thermometer was used to monitor the temperature fluctuation during leaching. The temperature was found to change within ± 2.5 °C of the required temperature. For experiments at higher temperature, i.e. 35 to 55 °C, a condenser was used to minimise water and ammonia loss. The reactor was nearly a

closed system with only one open on the condenser. Water around 10 °C flew through the condenser. The pH values were also detected by a pH metre in the whole leaching process. Prior to the leaching, the ammonia carbonate was firstly dissolved into the ammonia solution and demineralised water was added to reach a liquid volume of the required liquid-to-solid ratio. The solution was kept in the reactor for a period to reach the required temperature before the ICT waste was added. Liquid samples were taken at certain time intervals to track the leaching behaviour of different metals in the ICT waste. The sample size was 1–2 mL which is considered to have an insignificant effect on the liquid-to-solid ratio. Since the waste exhibits high heterogeneity, the residue after leaching was also analysed to diminish any discrepancy in composition. The liquid solution was analysed by induction coupled plasma-optical emission spectrometry (ICP-OES, PerkinElmer Optima 3000DV) and the residue was analysed by XRF. The extracted fraction of different metals is therefore calculated from

$$X_M = W_t / (W_t + W_r) \quad (1)$$

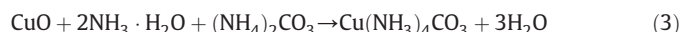
where W_t and W_r are the metal contents in the sampled solution at time t and the final solution, respectively; W_r is the metal content in the final residue after leaching.

2.3. Characterisation

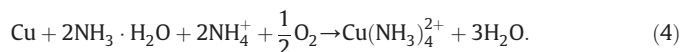
A sequential wavelength dispersive X-ray Fluorescence Spectrometry (XRF, PANalytical Axios) was used for compositional analyses of the bulk composition and the composition of the residue after leaching. The sample prepared according to the procedures in Sun et al. (2015) was pressed into pellets for analyses. The morphologies of the ICT waste were both characterised with a digital camera and scanning electron microscope (SEM, JEOL JSM 6500F) with energy-dispersive spectroscopy (EDS). The sample was polished and carbon coated when necessary. Liquid samples were diluted and prepared for analyses by ICP-OES.

3. Results and discussion

In this process, the recovery of copper from the ICT waste is based on ammonia leaching under ambient condition (room temperature and air pressure 1 atm with 20.9 vol.% oxygen). Ammonia leaching has demonstrated significant selectivity for copper recovery from such waste (Xiao et al., 2013). By simultaneously bubbling air into the leaching solution as shown in Fig. 1, copper is firstly oxidised by dissolved oxygen at the surface of the copper metal (Eq. (2)). The freshly formed copper oxide is further dissolved by the ammonia leaching solution (Eq. (3)).



The total reaction can be expressed by



It can be realised that the copper extraction is a complex gas–liquid–solid process. After dissolution of oxygen (air) into the ammonia solution and the reaction of possible polymeric insulation at the surface of copper metal, the leaching of copper includes the following stages: i) diffusion of oxygen and ammonia/ammonia salt from the bulk solution to the copper metal surface through the liquid boundary layer; ii) chemical reaction (4) at the surface; and iii) diffusion of ammine complex from the metal surface to the bulk solution. The copper extraction can be controlled by either of the steps or a combination of several steps. In order to reach a better understanding of the copper extraction

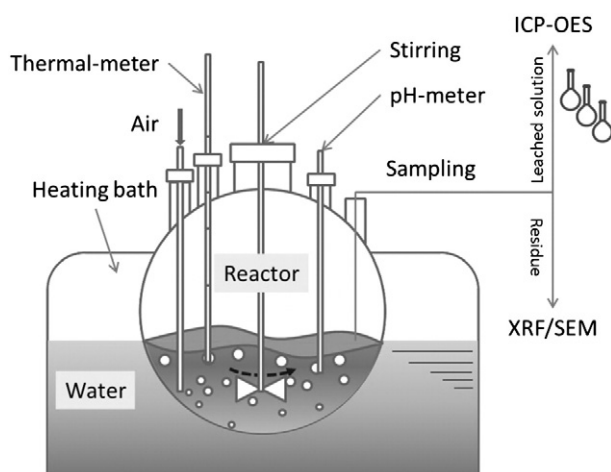


Fig. 1. Schematic plot of the leaching set-up.

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