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Effective treatment for electronic waste - Selective recovery of copper by combining electrochemical dissolution and deposition



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ABSTRACT

The difficulties for selective metal recovery from electronic waste are induced by the high complexity of such waste stream and complicated purification procedures. In this research, an electrochemical stimulated method is demonstrated to selective recover copper from an industrially provided electronic waste. More specific, one-step selective copper recovery was achieved by combining electrochemical dissolution and deposition in ammonia-based electrolyte. In the process, copper instead of other impure metals from the electronic waste is selectively dissolved and electrochemically enhanced at the anode. On the other side of the cell, pure copper was electrodeposited at the cathode. The current efficiency was found to be significantly influenced by the applied current density if galvanostatic process was facilitated. Both temperature and copper concentration in the electrolyte were found to have positive effects on the current efficiency. In a well-controlled process, copper of 99.56 wt% purity can be obtained which can be further improved by preventing potential oxidation on the electrodeposited layer. This research provide a possibility for selective copper recovery in one-step by substantially decreasing the purification cost and simplifying copper recovery procedures from electronic waste.

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1. Introduction

Waste electric and electronic equipment (EEE) has been increasing substantially worldwide in the last decades. It is considered to be one of the fastest growing solid waste streams and increases at a rate of around 10 million tons in the EU (Henckens et al., 2014; Nelen et al., 2014) with a world increasing rate of 20–50 million tons annually (Sun et al., 2017). Since a number of WEEE streams are classified as hazardous waste, its management and recycling is an important issue of worldwide environmental concern (Henckens et al., 2014; Nelen et al., 2014). On the other hand, significant content of valuable metals, including copper, silver, gold, makes WEEE a strategically important secondary resource. According to the new WEEE directive issued in 2012, WEEE export has been clearly forbidden (EU, 2012). However, in accordance to the current technological situation, a large number of WEEE streams are not properly treated with a low recycling rate

globally. The effective recycling rate (average) in the EU is only 37% in 2013 and 28% in China in 2015 according to the up to date reports although the rate is not in line with the management targets and some EU member countries mitigate higher recycling rates than the others (Baldé et al., 2015; Zhang, 2016). There is still significant room to be improved. With the recycling rate, a large fraction of the recycled WEEE is with relative high-quality which means small fraction of less valuable metals and high content of valuable metals (e.g. low value Fe comparing with high value Cu), insignificant compositional variation and stable material supply. When the WEEE is low-quality with low quantity of valuable metals, complex composition and phase contents, the recycling becomes technically difficult with high processing cost and potentially induces significant secondary pollution.

In literature, the processing technologies can be categorised into three types, i.e. pyrometallurgy, hydrometallurgy and biometallurgy. Prior to metallurgical processing, a variety of physical/ mechanical processes need to be applied to preliminarily separate end-of-life electronic products into a number of streams (Kahhat and Williams, 2009; Nnorom and Osibanjo, 2008; Widmer et al., 2005). In line with the compositions, materials can be further



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extracted via aforementioned metallurgical processes. A typical metallurgical process of WEEE recycling is based on the principle of pyrometallurgy which needs to burn the organic materials at high temperature, smelt/reduce the targeting metal and oxides. After further being refined or physio-chemically treated, metallic or salt products can be recovered (Bigum et al., 2012; Flandinet et al., 2012; Ni et al., 2012; Pareuil et al., 2010; Tange and Drohmann, 2005; Tuncuk et al., 2012).

A hydrometallurgical process is usually near to room temperature or a high temperature under pressure and it is considered to be potentially effective if WEEE is with complex composition and minor calorific value (Fleming, 1992; Gao et al., 2017; Ha et al., 2010; Havlik et al., 2011; Jha et al., 2001; Kim et al., 2011; Shuva and Kurny, 2013; Sun et al., 2015b). In the process, shredding and leaching are commonly introduced and the targeted elements together with the impurities are usually leached into a water based solution. With further purification and separation of the impurities, products can be further recovered. In a typical hydrometallurgical process, it is portable to facilitate metal recovery even with low concentrations through a range of technologies, e.g. sedimentation, solvent extraction and recrystallisation. Although it still needs to include a step of waste water treatment and the productivity is usually lower comparing with a pyrometallurgy process (Auer et al., 1998; Cui and Zhang, 2008; Reck and Graedel, 2012; Sum, 1991), it is still considered to be a technology of flexible, high recovery selectivity of critical metals and potentials of eliminating secondary waste (e.g. waste water). On top of the technological issues of electronic waste recycling, its effective management is also of great importance. With a proper model, the criticality and recyclability of different metals in electronic waste can also be identified (Sun et al., 2016a; Zeng and Li, 2016). It can be noticed that copper as one of the main values in electronic waste constitutes the importance for an electronic waste stream being considered for effective recycling.

Copper is among the most important/valuable metals in WEEE and it is commonly found in both mono-streams or relatively 'clean' WEEE, for instance, copper wires, printed circuit boards (PCB) (Cucchiella et al., 2016), waste mobile phones (Zhang and Xu, 2016), and complex electronic waste, such as shredding residue of mixtures of different end-of-life products or waste from different streams (Sun et al., 2015b). Hydrometallurgical process to recover copper usually includes steps of leaching and subsequent electrowinning (Diaz et al., 2016). The efficiency of copper recovery are highly influenced by the implemented conditions in both steps. In the leaching step, mineral acids were frequently introduced while base metals including iron, aluminum, lead etc. were all leached into the solution. Purification or solvent extraction had to be applied in order to obtain a solution ready for electrowinning (Gao et al., 2016). Direct electro-dissolution and electrowinning was tried by using ball-milled powder from waste PCB and HCl-based electrolyte solution was used to recover copper. However, high energy consumption from ball-milling and insufficient electric contact of the PCB powder which require additional effort to prepare suitable electrode were noticed (Guimarães et al., 2014). Meanwhile, it is not suitable for processing complex electronic waste. Therefore, effective copper recovery from the complex waste is still a challenge due to high content of other metals by using a hydrometallurgical process. Recently, an ammonia-based process has been developed in accordance to the treatment of such waste where copper could be selectively extracted in the leaching step while the leach solution is subjected to electrowinning in order to obtain pure copper (Sun et al., 2016b). In this research, we demonstrate a process for one-step selective copper recovery by combining these two technologies, where direct electro-leaching and -winning is facilitated.

2. Experimental

2.1. Waste sample preparation

The complex WEEE is provided by Van Gansewinkel Groep (VGG) and was characterised in accordance to the method developed in (Sun et al., 2015a). It contains a large fraction of sand, stone, glass, ceramics and plastic particles. Except copper, it contains a range of other base metals. Most metallic items are coated with different metallic or non-metallic materials and some wires are encapsulated in plastic cables or embedded between plastic boards. These diverse conditions may highly decrease the degree of liberation and the extraction efficiency. The material was shredded into size of less than or equal to 8 mm (Sun et al., 2015a). This size is defined to be a limit since further shredding will significantly increase the processing costs. A typical view of the waste after pre-treatment is presented in Fig. 1 and the average composition is given in Table 1.

2.2. Electrochemical process

The electrochemical recovery process was performed in a specially designed PMMA (Poly-methyl methacrylate) cell with a wall thickness of 5 mm and dimensions of 100 \times 70 \times 100 mm $(L \times D \times H)$ which results in a gross volume of 0.7 L. The plates were laser-cut and melted together with chloroform. The experimental set-up is shown in Fig. 1. The complex electronic waste was pressed via a stainless steel cylinder die with a pressure of 40 MPa in order to prepare pellet samples of around 2~3 g. The sample was subsequently introduced into an anode basket (Titanium) with a mesh size of 1.5 mm \times 1.5 mm (Fig. 1). As it is important that the basket does not dissolve in the electrolyte, the titanium basket was plated with platinum. As the cathode, a copper plate was used, from which one side was made inert by covering with resin materials during the process. The copper electrode had a height of 100 mm and a width of 25 mm (the emerging area is 25 mm \times 25 mm). The electrodes were weighted before and after each test. The electrolyte had a composition of 12 wt% ammonia and 155 g/L ammoniumsulphate. Agitation was in some cases provided by electrolyte stirring with a magnetic stirrer to improve mass transport in the solution. Electrochemical experiments were carried out with a potentialstat/galvanostat (VersaSTAT4, Princeton Applied Research) interfaced with a PC running software (VersaStudio 2.43.3). The reference electrode in this research was a leak-free Ag/ AgCl electrode ($E^0 = 0.222$ V vs. SHE, 3M KCl). The pH of the electrolyte was measured during the experiment with the WTW inolab pH 7310. Composition of the liquid samples were diluted and prepared for analyses with induction coupled plasma-optical emission spectrometry (ICP-OES, PerkinElmer Optima 3000DV). 3D optical microscope (Kevence, VHX-5000) and scanning electron microscope (SEM, JEOL JSM 6500F) with energy-dispersive spectroscopy (EDS) were used for morphology characterisation when needed.

3. Results and discussion

3.1. Electrochemical behavior of the electrodes

As aforementioned, the pellet of electronic waste was used as the anode to experience electrochemical dissolution and to directly recover copper at the cathode. It is therefore important to understand the electrochemical behavior of the anode especially when the pellet is introduced. One of the important feature is the dissolution potential with which the thermodynamics can be reflected. The anodic reactions can be given as Download English Version:

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