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An integrated and environmental-friendly technology for recovering valuable materials from waste tantalum capacitors

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ABSTRACT

Waste printed circuit boards (WPCBs) are valuable urban ore for recycling. Many efforts have been done to recover resources from basal boards of WPCB. In addition to basal boards, WPCBs contain many electronic components. However, the recycling technology for electronic components has been poorly developed. This study proposed an integrated and environment-friendly technology for recycling waste tantalum capacitors (WTCs) of WPCBs. Firstly, the WTCs were treated by argon pyrolysis to decompose mold resin. The pyrolysis temperature of 550 °C and holding time for 30 min were considered as the optimal parameters. Then, the pyrolysis residues were performed by crush and magnetic separation to recover nickel-iron terminals. Finally, chloride metallurgy (CM) was used to recover rare metal tantalum (Ta). The Ta recovery rate could reach 92.87 ± 0.36%. The optimal parameters were determined as temperature of 493 °C, adding $FeCl_2 \cdot 4H_2O$ of 53 wt% and holding time for 150 min based on the response surface methodology (RSM). This study exploits an efficient approach for recycling WTCs and provides a reference for future industrial application.

1. Introduction

With the development of science and technology, the number of electronic products has dramatically increased in the last two decades. Meanwhile, technological innovation and intense marketing shorten the life cycle of electronic products, resulting in the generation of large amounts of e-waste (Ghosh et al., 2015). It is estimated that nearly 45 million tons of e-waste are discarded globally per year, and the number is growing exponentially (Prabaharan et al., 2016). Printed circuit boards (PCBs), the integral parts of any electronic products, are particularly rich in metallic materials, including base, precious and rare metals (Zhang and Xu, 2016). The concentrations of these metals in PCBs are much higher than their respective rich-content minerals, which make waste PCBs (WPCBs) economically attractive urban ore for recycling (Li et al., 2015).

During the recycling process of WPCBs, electronic components (ECs), mounted on the WPCBs, were usually firstly dismantled. Then the waste printed wiring boards (WPWBs, WPCBs without ECs) were undergone the resource recovery process (Wang and Xu, 2015). Some advance technologies such as integrated mechanical-physical methods (Li et al., 2007; Li and Xu, 2010), hydrometallurgy (Havlik et al., 2010; Kim et al., 2011) and supercritical water (Li and Xu, 2015; Xiu et al., 2013; Liu et al., 2016) have been successfully applied to recover resources from WPWBs. However, the recycling technologies for ECs were poorly developed. Actually, WPCBs contain various ECs such as chips, resistors, transistors and capacitors (ceramic, aluminum and tantalum). Among them, tantalum capacitor (TC) is present in almost all electronic products owing to its advantage of small volume, large capacity and high thermal stability. For example, the PCB of mobile phone (3G technologies), notebook (2 GHz) and digital camcorder respectively contains about 36, 22 and 13 of these special capacitors (Angerer et al., 2013). Moreover, TC contains about 45 wt % of rare metal tantalum (Ta), and other valuable metals such as Ni (nickel) and iron (Fe), so it is attractive for recycling (Mineta and Okabe, 2005).

However, recovering valuable materials from WTCs is difficult because of the tightly covered mold resin, as presented in Fig. 1. The mold resin consists of silica, epoxy resin, phenolic novolac resin and flame-retardants (Katano et al., 2014; Iji, 1995). If WTCs were not disposed or recycled properly, the hazardous organics will be released into the environment, and then causing serious environmental pollution. So far, several related research such as combustion (Mineta and Okabe, 2005; Fujita et al., 2014), steam gasification with NaOH (Katano et al., 2014), chemical treatment (Mineta and Okabe, 2005; Spitczok von Brisinski1 et al., 2014) and phase

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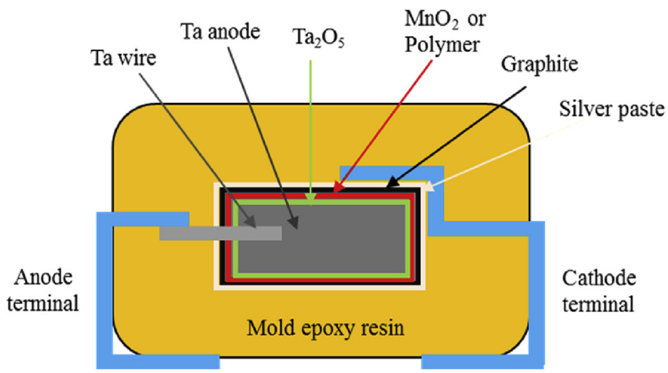

Fig. 1. Schematic illustration of a TC.

separation (Kikuchi et al., 2014), have been done to remove the organics and recover valuable materials from WTCs.

Pyrolysis is a quite promising technology for recycling organics compared with combustion and solvent leaching (Chen et al., 2010; Tripathi et al., 2016). On one hand, pyrolysis is conducted without oxygen, so the organics are decomposed to oils and gases, which can be recycled as fuel or feedstock; on the other hand, pyrolysis tends to be more effective than solvent leaching. Therefore, pyrolysis can be used as a pretreatment to recycle the organics of WTCs. However, the pyrolysis characteristic of the organics in WTCs has not been investigated. Besides, the recovery of valuable metals, especially rare metal Ta, is a critical process. Since Ta is a high-melting and corrosion-resistant metal (melting point: 2995 °C, insoluble in aqua regia), the recovery of Ta is usually achieved by dissolving or smelting the other components (Mineta and Okabe, 2005; Kikuchi et al., 2014). Consequently, large amounts of energy and chemicals were consumed during the recycling process. Besides, the obtained Ta had a low-grade. Chloride metallurgy has been proved to be an effective technology to extract rare metals from their ores and metal wastes (Jena and Brocchi, 1997; Kanari et al., 2009; Ma et al., 2012). In the CM process, valuable metals are converted to their corresponding chlorides and then separated based on their different volatility between the metal chlorides (Jena and Brocchi, 1997). This process does not produce liquid waste and is suitable for industrial production. In the previous study, we investigated the fundamental principles of chloride metallurgy (CM) for extracting Ta from WTCs. The results suggested that Ta could selectively react with $FeCl_2$ and the generated $TaCl_5$ can be easily separated and then condensed in the condensation zone. As a result, tantalum oxide with 99% purity could be obtained (Niu et al., 2017). However, the optimized parameters for industrial application were lacking.

Based on above considerations, this study proposed an integrated process including argon pyrolysis, mechanical-physical separation and chloride metallurgy, as presented in Fig. 2. The pyrolysis characteristic of the organics from WTCs was investigated. The CM process was optimized by applying central composite design (CCD) under response surface methodology (RSM) for industrial application. In addition, Ni-Fe terminals were also recovered by mechanical-physical separation. This study aims to exploit an effective and environment-friendly process for the maximum recovery of WTCs.

2. Materials and methods

2.1. Materials

The WTCs used in this study were obtained by an automatic disassembly system, as presented in Fig. 3. The major composition of the WTCs was listed in Table 1. $FeCl_2 \cdot 4H_2O$ (99.5%, Aladdin) was chosen as the chlorinating agent, argon (Ar, 99.99% purity) was used as the shielding gas.

2.2. Apparatus

The pyrolysis and chloride metallurgy (CM) experiments were conducted in a quartz tube furnace, as shown in Fig. S1 of Supporting Information (SI). The main body consisted of a body of

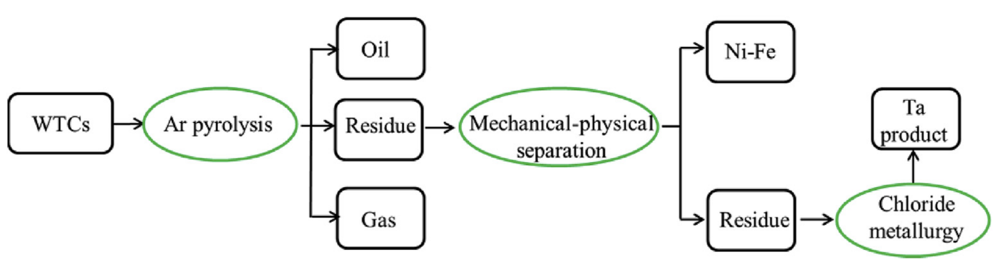

Fig. 2. Flow sheet of recycling process for WTCs.

Fig. 3. An automatic system for WTCs disassembly from WPCBs.