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Comprehensive process for the recovery of value and critical materials from electronic waste

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ABSTRACT

The development of technologies that contribute to the proper disposal and treatment of electronic waste is not just an environmental need, but an opportunity for the recovery and recycle of valuable metals and critical materials. Value elements in electronic waste include gold, palladium, silver, copper, nickel, and rare earth elements. This paper presents a technical assessment of the steps involved in a scheme that enables efficient recovery of value and critical materials from scrap mobile electronics. An electrochemical recovery process, based on the regeneration of ferric ion as a weak oxidizer, is studied for the selective recovery of base metals while leaving precious metals for separate extraction at reduced chemical demand. A separate process recovers rare earth oxides from magnets in electronics. Recovery and extraction efficiencies ca. 90% were obtained for the extraction of base metals from the non-ferromagnetic fraction in the two different solution matrices tested (sulfuric and hydrochloric acid). The effect of the pre-extraction of base metals in the increase of precious metals extraction, performed by means of anaerobic extraction in acid media, was assessed for the selective recovery of rare earths. A comprehensive flow sheet was developed to process electronic waste to value products.

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1. Introduction

Every year, billions of mobile electronic devices (e.g. cell phones, tablets, and laptops) are sold around the world. Because new technological developments emerge every day, the average time that a mobile electronic device is used before it is replaced by a newer model decreases. Most of these mobile devices can find a profitable market to a second life cycle, either as a refurbished or remanufactured product (Geyer and Doctori Blass, 2010). However, the exponential growth of the mobile electronics market, the increasing production of electronic waste (e-waste), and the reduced lifetime (Huang et al., 2009), vastly increase the need for the development of technologies for the recycling of end-of-life mobile electronic devices. Besides solving the environmental concerns associated with the landfill disposal and subsequent leaching of hazardous elements, such as brominated flame retardants, and heavy metals (Huang et al., 2009; Kiddee et al., 2013; Ghosh et al., 2015), the recycling of e-waste has an enormous potential as an

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http://dx.doi.org/10.1016/j.jclepro.2016.03.061 0959-6526/© 2016 Published by Elsevier Ltd. alternative source of value metals and critical materials (Lister et al., 2014; Hageluken, 2006; Sprecher et al., 2014). End-of-life mobile electronic devices possess a diverse composition of base and value metals, such as Cu, Ni, Pd, Au, and Ag, in concentrations that exceed those found in mineral ores (Ghosh et al., 2015; Ubaldini et al., 1998; Akcil et al., 2015). This scenario has activated a significant growth of the e-waste recycling industry in the U.S., processing over 4.4 Mt or e-waste in 2011 to produce commodity products such as steel and copper (ISRIINC, 2016). Mobile electronic devices also contain small, but significant, quantities of rare earth elements (REE) in speakers, vibrators, and hard disks, which can also be recovered and recycled (Buchert et al., 2012). The U.S. Department of Energy (DOE) has classified REE as critical elements for national energy security. Therefore, the recycling and recovery of REE from e-waste can contribute to the stabilization of the REE market and reduce the dependence of REE mining (Tukker, 2014; Bandara et al., 2014). Nevertheless, the need of cost efficient technologies for the recovery of REE is slowing the development of the REE recycle industry (Binnemans et al., 2013).

The distribution of value and critical elements in cell phones is presented in Table 1 (Geyer and Doctori Blass, 2010; Akcil et al., 2015; Buchert et al., 2012; Sullivan, 2006). An estimated

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L.A. Diaz et al. / Journal of Cleaner Production xxx (2016) 1-9

Table 1
Composition of value and critical material in cell phone

Metal	wt% Cell phone measured ^a	wt% Non-magnetic fraction ^a	wt% Ferromagnetic fraction ^a	wt% Cell phone literature
Cu	22.62	19.38	2.68	16 (Sullivan, 2006)
Sn	3.075	1.95	_	0.61 (Geyer and Doctori Blass, 2010)
Zn	_	1.40	0.60	0.59 (Geyer and Doctori Blass, 2010)
Pb	_	0.17	0.006	0.3 (Akcil et al., 2015)
Fe	_	0.94	78.78	5 (Akcil et al., 2015)
Ni	13.49	0.82	6.84	0.1 (Akcil et al., 2015)
Ag	0.26	0.3	_	0.3 (Buchert et. al., 2012)
Au	0.053	0.06	_	0.03 (Buchert et. al., 2012)
Pd	0.0273	0.01	_	0.01 (Buchert et. al., 2012)
Pr	0.0095	_	0.11	0.01 (Buchert et. al., 2012)
Nd	0.0818	_	0.90	0.05 (Buchert et. al., 2012)
Dy	0.0137	_	0.10	_

^a Experimental values obtained after digestion in aqua regia and/or 50% HNO₃.

percentual contribution of metals and rare earth elements (REE) to the total recoverable value of the e-waste, based on the average value of metals during 2014 (ca. USD 3.75 per cell phone unit and ca. USD 7.33 per laptop unit), is shown in Fig. 1 for cell phones (a) and laptops (b). It is evident that the highest recovery value comes from metals with the lowest concentration. A small percentage of gold is found in mobile electronics, and yet it contributes to the highest portion of the total recoverable value of e-waste. Based on this fact, extensive research is currently being performed in the development and optimization of hydrometallurgical, non-cyanide, processes focusing on the recovery of value metals (Birloaga et al., 2013; Ha et al., 2014; Kim et al., 2011a; Yang et al., 2011; Behnamfard et al., 2013; Tuncuk et al., 2012).

Hydrometallurgical treatment of e-waste has been found to be more efficient and environmentally friendly than other recovery processes, such as physical separation and pyro-metallurgical processes (Ghosh et al., 2015). With a hydrometallurgical approach, the low leaching selectivity and the low concentration of precious metals, relative to the total metal content, lead to a significant use of chemicals in the dissolution of the most abundant metals, such as Cu or Sn (Ghosh et al., 2015; Birloaga et al., 2014; Sun et al., 2015).

Electrochemical methods have been developed to reduce the use of reagents (Ghosh et al., 2015). Electrochemically generated chlorine (Cl₂) is a strong oxidizer capable of dissolving most metals including gold. However, the leaching rate is limited by the solubility of Cl₂ (Kim et al., 2010, 2011b). Moreover, the use of Cl₂ as an oxidizer presents issues regarding worker safety and corrosion of materials used in construction. An alternative electrochemical recovery (ER) process, already tested for the efficient recovery of metals from ores at low temperatures (Casas et al., 2008; Sandberg and Huiatt, 1986), is based on the use of a weak oxidizer (Fe³⁺), which selectively dissolves the less noble metals in the e-waste (Equation (1)) (Lister et al., 2014; Fogarasi et al., 2014). A

thermodynamic analysis of the oxidation conditions required for the extraction of metals, such as Cu, Sn, and Ag, using Fe^{3+} as the oxidant was presented in a previous work (Lister et al., 2014).

In the ER process, the oxidizer is regenerated at the anode of an electrochemical cell (Equation (2)), coupled with the electrowinning of metals in the cathode (Equation (3)).

$$n\mathrm{Fe}^{3+} + M \to M^{n+} + n\mathrm{Fe}^{2+} \tag{1}$$

$$Fe^{2+} \rightarrow Fe^{3+} + e^{-}, \quad E^0 = 0.771 \text{ V vs. SHE}$$
 (2)

$$M^{n+} + ne^- \to M \tag{3}$$

where *M* is the metal leached, *n* is the metal-ion charge, and E^0 is the standard reduction potential vs. standard hydrogen electrode (SHE).

Up to 50% energy reduction, compared to the current copper electrowinning processes (Beukes and Badenhorst, 2009), can be achieved just by the substitution of the oxygen evolution reaction ($E^0 = 1.229$ V vs. SHE) with the oxidation of Fe²⁺ to Fe³⁺ in the anode during the electrowinning of base metals. The main advantage of this process is that it increases the concentration of gold and palladium in the waste material by the selective and complete removal of the base metals with minimal chemical input.

The ER process has shown promising results in terms of having a low environmental impact (Fogarasi et al., 2013), low energy consumption (Fogarasi et al., 2014), and high base metals recovery, while leaving Au and Pd intact for removal in separate stages (Lister et al., 2014). Nevertheless, more research is required to improve the leaching kinetics, increase the solid to liquid ratio (S/L) in the leaching treatment, and assess the effect of the base metals extractions in the enrichment and recovery of precious metals.

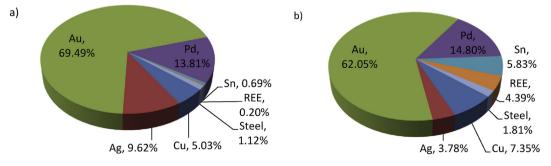


Fig. 1. Value distribution of metals and critical materials in a) cell phones and b) laptops.

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