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Research article

Physical and thermal processing of Waste Printed Circuit Boards aiming for the recovery of gold and copper

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ABSTRACT

The recovery of electronic waste to obtain secondary raw materials is a subject of high relevance in the context of circular economy. Accordingly, the present work relies on the evaluation of mining separation/concentration techniques (comminution, size screening, magnetic separation and gravity concentration) alone as well as combined with thermal pre-treatment to recover gold and copper from Waste Printed Circuit Boards. For that purpose, Waste Printed Circuit Boards were subjected to physical processing (comminution, size screening in 6 classes from < 0.425 mm to > 6.70 mm, magnetic separation and gravity concentration) alone and combined with thermal treatment (200–500 °C), aiming the recovery of gold and copper. Mixed motherboards and graphic cards (Lot 1 and 3) and highly rich components (connectors separated from memory cards, Lot 2) were analyzed. Gold and copper concentrations were determined before and after treatment. Before treatment, concentrations from 0.01 to 0.6 % wt. and from 9 to 20 % wt. were found for gold and copper respectively. The highest concentrations were observed in the size fractions between 0.425 and 1.70 mm. The highest copper concentration was around 35 % wt. (class 0.425–0.85 mm) and when analyzing memory card connectors alone, gold concentrations reached almost 2% in the same class, reflecting the interest of separating such components. The physical treatment alone was more effective for Lot 1/3, compared to Lot 2, allowing recoveries of 67 % wt. and 87 % wt. for gold and copper respectively, mostly due to differences in particles size and shape. The thermal treatment showed unperceptive influence on gold concentration but significant effect for copper concentration, mostly attributed to the size of the copper particles. Concentrations increased in a factor of around 10 when the thermal treatment was performed at 300 °C for the larger particles (1.70–6.70 mm); the best results were obtained at 400 °C for the other sizes, when the highest rate of thermal decomposition of the material occurred.

1. Introduction

The constant search for technological solutions to simplify daily routine tasks has created a new environmental problem - the electronic waste - and, particularly, the waste of printed circuit boards (WPCB), associated to most electronic products. The amount of electronic waste generated might be determined taking into account the products put on the market and the appropriate lifespan (probability of product discard over time). Based on that, in 2014 the production of 9.5 Mt was estimated and around 10.4 Mt of electronic waste are predicted for 2024 (EC-UNU, 2014). The most recent Eurostat data predicts around 9.8 Mt of products put on the market in the year 2015. The recovery of electronic waste to obtain secondary raw materials is fundamental to reduce the amount of waste sent to landfill, decreasing the associated environmental impacts and allowing the development of a more sustainable society (Ghosh et al., 2015; Hadi et al., 2015; Huang et al., 2009; Sthiannopkao and Wong, 2013).

WPCB are mostly composed by organic materials (up to 30%), ceramics (up to 30%) and metals (up to 40%) (Ogunniyi et al., 2009; Yuan et al., 2007; Zeng et al., 2012). Their importance, from an economic and environmental point of view, relies in the metals present (high concentration and purity), and in the hazardous nature of some of its constituents (Choubey et al., 2015; Ortuño et al., 2014; Tuncuk et al., 2012; Zeng et al., 2012). WPCB may contain about 250 g/t of gold and 20 % wt. of copper, very high values when compared with the gold ores with concentrations between 1 and 10 g/t, and copper ores with grades between 0.5 and 1 wt% (Tuncuk et al., 2012).

Highly toxic elements like lead and bromine, known to cause irreversible damages to the environment and human health, are used in the solders and as part of the flame retardants (Jianzhi et al., 2004). Taking into account environmental and health safety concerns, the EU directive 2002/95/EC restricted the use of lead, mercury, cadmium,

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hexavalent chromium and two flame retardants added to plastics: polybrominated biphenyls (PBB) and polybrominated diphenylethers (PBDE) (Sthiannopkao and Wong, 2013; Tuncuk et al., 2012; Xuefeng et al., 2005).

Most of the research work on WPCB focus on the recovery of the metals present, using pyrometallurgical, hydrometallurgical and biometallurgical technologies, as well as mechanical/physical processes. Pyrometallurgical technologies use thermal processes for the recovery, requiring large amounts of energy and causing the emission of toxic compounds such as dioxins, furans, polybrominated organic pollutants and polycyclic aromatic hydrocarbons in gaseous effluents, difficult and expensive to treat (Rocchetti et al., 2013). Hydrometallurgical technologies use leaching agents such as cyanide, thiourea, thiosulfate (sodium and/or ammonium), aqua regia and halides (chlorine, bromide and iodide) to dissolve the metals present in WPCB followed by solvent extraction, precipitation or electrometallurgical processes for metals recovery; such processes generate highly acidic or other toxic liquid effluents which require adequate management, with high associated environmental and economic impacts (Duan et al., 2011; Ghosh et al., 2015; Hadi et al., 2015; Silvas et al., 2015; Zhang et al., 2012). Biometallurgical processes take advantage of the capacity of some microorganisms to absorb/adsorb the metals, combining their metabolic necessities with the chemical transformations of the minerals, which might be a simple and efficient way for their recovery; the slow microbial growth and hence the very long times for the recovery are still significant disadvantages that difficult its application (Morin et al., 2006; Xiang et al., 2010; Yang et al., 2009).

Physical/mechanical processes take advantage of the physical properties of different materials (density, conductivity, magnetic permeability and ductility) to separate and concentrate them (Ghosh et al., 2015). Grinding or comminution is the key and transversal step for metals recovery regardless the used process, followed by grain size analysis (granulometric classification). Veit et al. (2006, 2002, 2005) performed different studies where the WPCB were comminuted into different particle sizes, and afterwards the metal fraction was concentrated using properties such as density, magnetic permeability and conductivity, as well as a combination of these with electrometallurgical processes, specifically electro-winning. Approximately 80% of the metals were concentrated in the coarse fraction (0.50–1.0 mm), with copper being the largest constituent (> 50 % wt). Guo et al. (2011) showed the importance of comminution in the liberation of the metal embedded in the epoxy matrix of the board, with the highest liberation occurring in the particles size range of 0.15–1.25 mm. Long et al. (2010) studied a combined process of pyrometallurgical and physical separation (density). The WPCB were cut and pyrolised (up to 550 °C at a heating rate of 10 °C/min with a holding time of 120 min), the ashes were then fragmented using a universal crusher and screened by sizes. Separation was performed through a vertical zigzag airflow obtaining an amount of copper close to 99 % wt. for the particles size range of 0.45–4.0 mm.

The new technological approaches, combining physical/mechanical treatments with thermal pre-treatments (Long et al., 2010; Zhou et al., 2010) and the application of mining separation/concentration techniques (Ventura et al., 2014) to recover metals from WPCB might be considered a more sustainable path for WPCB recovery. Their operational simplicity, together with minor alterations to the existing technologies, contribute for a broader implementation of such processes, potentially reducing environmental, social and economic costs associated with the incorrect waste management and the metallurgical processes further applied. In fact, very few studies are reported under this matter, which makes relevant the development of research with such focus. The present work relies therefore on the evaluation of mining separation/concentration techniques alone as well as combined with thermal pre-treatment to recover gold and copper from WPCB. In order to explore raw-material specificities, since studies performed report essentially the used of non-segregated wastes, the studies were performed with segregated waste, using three lots, consisting on motherboards and graphic cards and RAM cards with and without connectors.

2. Material and methods

In the case of Physical processing alone, the following steps were conducted: (1) Manual removal of bulkier and toxic components; (2) Cutting into smaller parts; (3) Comminution; (4) Classification into 6 sizes classes; (5) Magnetic separation; and (6) Gravity concentration.

In the case of Physical and Thermal processing, the following steps were conducted: (1) Manual removal of bulkier and toxic components; (2) Cutting into smaller parts; (3) Thermal treatment; (4) Comminution; (5) Classification into 6 sizes classes.

Each of the mentioned steps are described in more detail in the following subsections.

2.1. Dismantling and comminution

The WPCB used in this study were collected from obsolete personal computers of the Faculty of Engineering of the University of Porto (FEUP), and consisted on motherboards, graphic cards, modem cards and memory cards, of different brands, models and years. The original computers were from the decade 1998–2008 and most of them were from near the year 2000.

In order to study the physical processes, WPCB were divided into two lots. The first one (Lot 1), containing motherboards and graphic cards, was divided into two fractions (replicas), whereas the second one (Lot 2), containing modem and memory cards, was divided into two fractions, one containing modem cards and memory cards without the connectors (Fraction 2.1) and other containing only their connectors (Fraction 2.2). Finally, a third lot (Lot 3), with similar composition as Lot 1, was used to study the thermal/mechanical process, being divided into four fractions, which were submitted to the different evaluated temperatures. The sampling process is depicted in Fig. 1.

All the WPCB were manually stripped from their bulkier and toxic components (e.g. metallic brackets for support in the CPU; CMOS – complementary metal-oxide-semiconductor backup battery) and cut into smaller parts, of about 8 cm × 5 cm (Lot 1 and 2) and 10 cm × 15 cm (Lot 3), using a manually operated metal hand shear.

Taking into account the amount of material present in the different lots, the fractions were comminuted (before physical treatment for Lot 1 and 2 or after thermal treatment for Lot 3) using either an ERDWICH single shaft shredding system M400/I-200 (Lot 1 and Fraction 2.1), or a RETSCH SM 200 (Fraction 2.2 and Lot 3). After comminution, the fractions were classified according to size using a vibrating sieve shaker (RETSCH AS 200) and the following nominal sieves openings: 6.70 mm, 3.35 mm, 1.70 mm, 0.850 mm and 0.425 mm, leading to the following 6 sizes classes: < 0.425, 0.425–0.850 mm, 0.850–1.70 mm, 1.70–3.35 mm, 3.35 mm–6.70 mm, and > 6.70 mm.

2.2. Physical/mining concentration processes

Lots 1 and 2 were subjected to magnetic separation followed by gravity concentration. The magnetic separation was performed to clean the material flow from ferromagnetic materials for further separation of the non-ferromagnetic metals, including those in study: gold and copper, through gravity concentration.

2.2.1. Removal of ferromagnetic components

The material flow was divided in two streams: a ferromagnetic and a non-ferromagnetic stream. The samples with a particle size higher than 0.850 mm were submitted to manual magnetic separation (using a magnet) whereas the remaining fractions were separated using a laboratory induced roll magnetic separator, a high field intensity equipment (field strengths of up to 1.2 T attainable in the gap between feed