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A wet dismantling process for the recycling of computer printed circuit boards



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ABSTRACT

In this research a new process for dismantling the components of the waste printed circuit boards (WPCBs) was developed. This process uses water and aqueous solutions of sodium hydroxide at conditions above solder melting temperatures which not only prevents pollution problems, but also separates the multilayers of WPCBs. The results show that removal of components in the WPCBs was achieved at 280 °C for 15 min with a 1 M NaOH solution. At these conditions glass fiber, epoxy resin and copper layers were separated and the resultant solution showed low concentration of copper, zinc, lead and iron ranging from 23 to 43 mg/kg of WPCBs and high concentration of tin and aluminium of 8 g/kg and 61 g/kg of WPCBs, respectively. About 97% of tin and 99% of aluminium in this solution were removed by precipitation.

1. Introduction

A large volume of waste from electric and electronic equipment (WEEE) is generated worldwide annually, being the fastest growing waste stream in the last years, growing at a rate three times higher than average municipal waste (Estrada-Ruiz et al., 2016). In 2018, WEEE will grow to

49.8 million tonnes with an annual growth rate of 4-5% (Johnson et al., 2018). Printed circuit boards (PCBs) are an essential part of almost all electric and electronic equipment (EEE), and its percentage varies between 3% and 6% of the total constitution of EEE (Zhou and Qiu, 2010). The great demands in EEE coming from technology improvements and society development always seeking for the high-performance create the need for frequent replacement of PCBs and generates large quantities of WPCBs for disposal. In last years, the average rate of PCBs production increased by 8.7% (Guo et al., 2009; Huang et al., 2009). WPCBs contain a variety of hazardous substances and heavy metals, namely halogenated flame retardants, cadmium, lead, and other metals that, in order of a sound environmental point of view, should be adequately managed, preferably recycled (Kaya, 2016; Zhou and Qiu, 2010). PCBs are composed by metals (40%), polymers (30%) and ceramic (30%) components (Guo et al., 2009; Kaya, 2016). The variety of metals present in WPCBs, copper in particular, and precious metals in quantities even higher than in the ores, makes this residue particularly attractive from the point of view of recycling and a significant secondary metal resource.

Nevertheless, recycling of WPCBs is still limited due to the heterogeneity and complexity of its components (Awasthi and Li, 2017; Duan et al., 2011; Jha et al., 2012). The recycling techniques for these materials include mechanical, thermal and chemical processing, or a combination of these processes (Birloaga et al., 2014; Cui and Zhang, 2008; Wang et al., 2017). The electric components (ECs) removal and separation of metal to the non-metal fraction from WPCBs is the most important step from the environmental and economic point of view. Currently, the ECs removal is performed manually or destroyed by mechanical or thermal pre-treatments. In manual dismantling chisels, hammers and cutting torches are used to open solder connections and separate ECs (Kaya, 2016). The PCBs are heated using electric heating plates, with exhausted gases collected, in order to melt and resell the chips and other ECs to acid strippers to further processing. This process causes a bad smell and a black fume, and for this reason is coming into disuse (Duan et al., 2011; Huang et al., 2009). The mechanical processes involve grinding, magnetic separation, size separation, air classification, density separation, corona electrostatic separation and eddy current separation. Using either physical or mechanical process, noble metals may be lost and emissions may arise from the dust generated during milling operations as well as from liquids used as transportation or separation medium.

Several studies have been carried out in the field of WPCBs recycling not only in the processes application but also in economic and environmental impact assessment (An et al., 2015; Neto et al., 2017). Wang et al. (2017) reported the researches of various authors that use

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the pyrometallurgical process to recycling of WPCBs. Yokoyama and Iji (1997) developed a technology to recycling WPCBs, where the ECs are removed by a combination of heating them to above the melting point of solder and applying such external forces as impact, shearing and vibration. Zhou and Qiu (2010) reported a new process to recover solder and organic materials from WPCBs using centrifugal separation and vacuum pyrolysis. The separation of solder from WPCBs was completed when they were heated at 240 °C and centrifuged at 1400 rpm for 6 min. According to Duan et al. (2011), the process of WPCBs dismantling using a hot fluid and chemical reagents has been reported in some studies. Wang and Xu (2017) studied the recovery of aluminium and iron from electrolytic capacitors from WPCBs. They combined heating treatment, crushing, sieving and magnetic separation to recover aluminium and iron from capacitors. They achieved a recovery of 97% of aluminium and 99% of iron, heating at 500 °C during 60 min. The major common point of dismantling is melting of solder to separate ECs from WPCBs, what is achieved for a temperature 40-50 °C higher than the melting point of solder. At these temperatures the production of pollutant gases may occur. In several studies found on recycling WPCBs, the focus is essentially on the recovery of noble metals (Awasthi and Li, 2017; Lu and Xu, 2017); and some of the technologies studied in ECs disassembly from WPCBs have low efficiency and high cost. In this context, developing a new and green technology for recycling WPCBs is essential. The main objective of this work was to study a combined process of dismantling and separation of the multilayers of the WPCBs using water or aqueous solutions of sodium hydroxide at conditions above solders melting temperatures.

The changes that occur in components were characterized and metals, halides and total organic carbon (TOC) were analysed in resultant solutions from this process. Additionally, due to the high content of aluminium and tin in the resultant solutions, a chemical treatment was carried out to treat them and recover these metals as hydroxides.

2. Materials and methods

2.1. Materials

The WPCBs used in all experiments were taken in obsolete computers from Faculty of Engineering of University of Porto. Following, the large components as universal serial bus (USBs) and high-definition multimedia interface (HDMIs) were removed. Due to the size of the reactor used at this work, the WPCBs were cut in pieces of 10×10 cm using a guillotine, following reduced in pieces of approximately 2×2 cm, with cutting scissors.

2.2. Methods

2.2.1. Chemical composition of WPCBs

The chemical composition of the WPCB samples was determined by X-ray fluorescence spectrometry analysis (XRF). To ensure the homogeneity of the samples for XRF analysis they were prior comminuted using a RETSCH SM 200 equipment and the resultant material was classified according to size using a vibrating sieve shaker (RETSCH AS 200) and the 1.0 mm and 0.5 mm sieves leading to the classes size of < 0.5 mm, 0.5–1.0 mm and > 1.0 mm. The samples for analysis were obtained using a Jones sampling equipment.

2.2.2. Differential scanning calorimetric analyses

The differential scanning calorimetric (DSC) analyses of WPCBs solder was performed using a Setaram equipment, model Labsys. The solder sample taken carefully from a piece of WPCB was placed in an aluminium crucible, heated at a rate of 10 °C/min up to 280 °C. A blank test was carried out with unloaded crucibles using the same conditions. The analyses were carried in duplicate and corrected with values of the blank test.

Table 1

Chemical methods for characterizing the solutions from the dismantling step.

Parameter	Methods
pH	4500 B – Electrometric Method (APHA, 1998)
Halides	4500 B – Argentometric Method (APHA, 1998)
Total Organic Carbon	EN 1484:1997
Total metals: Al, Sn, Cu, Zn, Fe, Ni, Pb	US EPA 7000B:2007 and AAS

2.2.3. Tests for WPCBs wet dismantling

The tests were performed in a Parr titanium alloy batch reactor with 450 mL having temperature control and pressure reading. All tests were carried out with stirring.

The experiments were performed in aqueous medium, i.e., only water or with either 0.1 M or 1 M NaOH solutions, that were previously prepared outside of the reactor in order to guarantee the total NaOH dissolution. The tests were carried out at temperatures of 220 °C, 240 °C, 260 °C or 280 °C and at different retention times. The selected temperatures were based on the result from the DSC analysis and all above the melting point of solder. Pressure ranged from 1.6 MPa to 6.0 MPa, this last in the tests at 280 °C. The samples used in the tests contained on average 6 pieces of WPCBs and had 10–20 g. The liquid/ solid ratios varied from 10:1 (w/w) to 20:1 (w/w).

2.2.4. Characterization of the solutions resultant from WPCBs wet dismantling

The chemical characterization of the solutions resulting from WPCBs wet dismantling was carried out according to the standards referred in Table 1. Metals quantification in the solutions was performed by Atomic Absorption Spectrometry (AAS).

3. Results and discussion

3.1. Chemical composition of WPCBs

Table 2 shows chemical composition of WPCB samples obtained by XRF concerning the main elements of interest. Zinc and gold present similar concentration in the 3 fractions analysed. Iron is the second highest concentration essentially in the lower size fraction. The opposite was verified for copper which has the lowest content in this fraction and the highest amounts are in sizes coarser than 0.5 mm; the similar results were obtained for the nickel. These results corroborate those obtained by Veit et al. (2006). Lead and tin were mainly at the sizes less than 1 mm. For all elements, the values found in WPCBs analyses are within the ranges presented in the literature, except for tin which has a lower content in these samples. Kaya (2016) reports values of 6-27% of copper, 0.2-2.2% of zinc, 0.3-5.4% of nickel, 1.0-4.2% of lead and 1.2-8.0% of iron. Chemical composition of PCBs is found in several studies (Birloaga et al., 2013; Cui and Zhang, 2008; Oishi et al., 2007; Veit et al., 2006; Yang et al., 2012) some of them based in studies from other authors (Stuhlpfarrer et al., 2016; Kaya, 2016; Silvas et al., 2015;

 Table 2

 Composition of the WPCBs obtained by XRF analysis.

Element, (%)	< 0.5 mm	0.5–1.0 mm	> 1.0 mm
Cu Z=	9.88 ± 0.26	17.93 ± 1.1	16.63 ± 3.24
Fe	0.38 ± 0.03	0.42 ± 0.09	0.38 ± 0.11
	3.21 ± 0.08	0.29 ± 0.05	0.22 ± 0.08
Ni	0.20 ± 0.01	0.42 ± 0.10	0.41 ± 0.06
Pb	0.36 ± 0.00	0.50 ± 0.09	0.06 ± 0.02
Sn	0.70 ± 0.01	0.90 ± 0.28	0.31 ± 0.08
Au	0.04 ± 0.00	0.04 + 0.01	0.03 ± 0.00
114		0101 = 0101	0.00 = 0.00

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