



Copper extraction from coarsely ground printed circuit boards using moderate thermophilic bacteria in a rotating-drum reactor



Michael L.M. Rodrigues^{a,*}, Versiane A. Leão^a, Otavio Gomes^b, Fanny Lambert^c, David Bastin^c, Stoyan Gaydardzhiev^c

^a Bio&Hydrometallurgy Laboratory, Department of Metallurgical and Materials Engineering, Universidade Federal de Ouro Preto, Campus Morro do Cruzeiro, Ouro Preto, MG 35400-000, Brazil

^b Centre for Mineral Technology – CETEM, Av Pedro Calmon, 900, 21941-908 Rio de Janeiro, Brazil

^c Mineral Processing and Recycling, University of Liege, Sart Tilman, 4000 Liege, Belgium

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ABSTRACT

The current work reports on a new approach for copper bioleaching from Printed Circuit Board (PCB) by moderate thermophiles in a rotating-drum reactor. Initially leaching of PCB was carried out in shake flasks to assess the effects of particle size ($-208\ \mu\text{m} + 147\ \mu\text{m}$), ferrous iron concentration (1.25–10.0 g/L) and pH (1.5–2.5) on copper leaching using mesophile and moderate thermophile microorganisms. Only at a relatively low solid content (10.0 g/L) complete copper extraction was achieved from the particle size investigated. Conversely, high copper extractions were possible from coarse-ground PCB (20 mm-long) working with increased solids concentration (up to 25.0 g/L). Because there was as the faster leaching kinetics at 50 °C *Sulfobacillus thermosulfidooxidans* was selected for experiments in a rotating-drum reactor with the coarser-sized PCB sheets. Under optimal conditions, copper extraction reached 85%, in 8 days and microscopic observations by SEM-EDS of the on non-leached and leached material suggested that metal dissolution from the internal layers was restricted by the fact that metal surface was not entirely available and accessible for the solution in the case of the 20 mm-size sheets.

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1. Introduction

Technological innovations have stimulated widespread utilization of electronic equipment, which has also resulted in a considerable increase in the generation of electronic waste (e-waste). Incineration and landfilling are often the ways to deal with electronic waste (Xiang et al., 2010), however such methods are increasingly being considered inappropriate because of their negative environmental impacts as shown by the recent studies (Brandl et al., 2001; Yang et al., 2009). Conversely, electronic wastes can also be viewed as alternative source of non-ferrous metals such as copper, nickel, and zinc as well as gold and silver (Lee and Pandey, 2012) and both pyrometallurgical and hydrometallurgical techniques can be applied for their recycling from e-wastes (Zhu et al., 2011).

Pyrometallurgy is particularly appropriate to recover copper and precious metals from some type of electronic waste such as

mobile phones, but the technique requires high grade feed and thus is not suitable for low grade e-wastes, including PCB. Moreover, pyrometallurgy is related to some environmental impacts such as greenhouse gases, formation of furans and dioxins and dust (Cui and Zhang, 2008; Ilyas et al., 2010; Zhu et al., 2013) and thus requires an off-gas treatment. In addition, rare earth metals, tantalum and gallium among other elements report to the slag phase because they are easily oxidized in the furnace atmosphere. Therefore, hydrometallurgy is considered an alternative because it is less complex and energy intensive and there is no generation of toxic gases (Tuncuk et al., 2012). Nevertheless, it is fair to admit that both approaches have their inconveniences and advantages making their combined use often the preferred scenario.

Bio-hydrometallurgy is a specialized branch of hydrometallurgy that utilizes microorganisms to solubilize metals and therefore is often viewed as eco-friendly process for the treatment of low-grade ores and wastes (Poulin and Lawrence, 1996; Watling, 2006). Although, this is one of the most promising biotechnologies in the metallurgical sector, limited research has been reported regarding the bioleaching of PCB and a few examples are

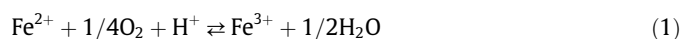
* Corresponding author.

E-mail addresses: mitchel.marques@yahoo.com.br (M.L.M. Rodrigues), versiane@demet.em.ufop.br, versiane.ufop@gmail.com (V.A. Leão).

Table 1
Bioleaching of copper from PCB.

Specific type and source of waste	Microorganisms	Leaching efficiency (%) and conditions	Type of reactor	References
PCB –208 μm + 147 μm	<i>Acidithiobacillus ferrooxidans</i> and <i>Sulfobacillus thermosulfidooxidans</i>	94% Cu extraction at 30 °C (<i>T. ferrooxidans</i>) and 99% at 50 °C (<i>S. thermosulfidooxidans</i>) in 6 days with 10 g/L Fe ²⁺ , 10 g/L of PCB and pH 1.75; 150 min ⁻¹	Shaking flask	Current work
PCB 20 mm-long	<i>S. thermosulfidooxidans</i>	85% Cu extraction in 8 days at 5 g/L Fe ²⁺ , 25 g/L PCB and pH 1.75. Rotating-drum reactor at 50 °C and 80 min ⁻¹	Rotating-drum reactor	Current work
PCB < 500 μm	<i>At. thiooxidans</i> and <i>At. ferrooxidans</i>	>90% Cu extraction in two-stages during 7 and 10 days at 5–10 g/L of PCB; 35 °C and 180 min ⁻¹	Shaking flask	Brandl et al. (2001)
PCB < 80 mesh	Genera <i>Acidithiobacillus</i> and <i>Gallionella</i>	95% of Cu in 5 days at 9 g/L of initial Fe ²⁺ , 20 g/L of PCB and pH 1.5; shaker at 30 °C and 120 min ⁻¹	Shaking flask	Xiang et al. (2010)
PCB 100–120 μm	<i>S. thermosulfidooxidans</i> and <i>Thermo-plasma acidophilum</i>	86% Cu extraction. Acid pre-leaching of 27 days and bioleaching of 280 days. Column reactor at 45 °C	Column reactor	Ilyas et al. (2010)
PCB 60–80 mesh	Enriched acidophilic bacteria	96.8% Cu extraction in 45 h; 12 g/L Fe ²⁺ , 12 g/L PCB and pH 2; shaker at 30 °C and 160 min ⁻¹	Shaking flask	Zhu et al. (2011)
TV circuit boards (STVB) < 250 μm	<i>At. ferrooxidans</i> , <i>L. ferrooxidans</i> and <i>At. thiooxidans</i>	35% Cu extraction in 90 h without external Fe ²⁺ and 89% Cu extraction at 8 g/L Fe ²⁺ at 10 g/L of PCB and pH 1.7; shaker at 35 °C and 170 min ⁻¹	Shaking flask	Bas et al. (2013)
PCB 50–150 μm	<i>S. thermosulfidooxidans</i> and <i>T. acidophilum</i>	85% Cu extraction in 18 days at 10 g/L of PCB; shaker at 45 °C and 180 min ⁻¹	Shaking flask	Ilyas et al. (2013)

summarized in Table 1. Iron-oxidizing bacteria produces ferric from ferrous iron (Eq. (1)), which can subsequently oxidize metals such as copper according to Eq. (2). In general, mesophilic bacteria belonging to the genus *Acidithiobacillus* and *Leptospirillum* are most commonly used in bioleaching.



In addition to these two bacterial genera thermophilic microorganisms may also be employed in bioleaching operations (Brandl et al., 2001; Pant et al., 2012). As a rule, leaching at elevated temperatures has the potential to result in more efficient and faster metal dissolution. In the bioleaching of copper sulfides for instance higher extractions with moderate thermophilies as compared to mesophiles were reported elsewhere (Pina, 2006; Schippers, 2007). Copper bioleaching from e-wastes with moderate thermophilies in shake flasks (Ilyas et al., 2007, 2013; Pina, 2006), stirred tanks (Ilyas et al., 2014) and lab scale columns (Ilyas et al., 2010) were also investigated. However, few studies have studied bioleaching of e-waste and printed circuit boards in particular using moderate thermophiles.

The reactors used in the bioleaching of sulfides are generally designed as stirred tank or air-lift type but concerning e-waste bioleaching there is a limited data about the type of reactor on process performance. A rotating-drum reactor could be suggested as alternative to the stirred-tank reactors with the potential advantage to treat material at increased pulp densities and reduce global energy consumption. It could be assumed that such a reactor will ensure reduced impact on the microbial cells due to the lower degree of collisions between the particles (Liu et al., 2007). Such configuration offers the possibility of using high solid loadings without negatively influencing the bio-oxidation of Fe²⁺ which tends to be the case when impeller-driven reactors are used (Jin et al., 2013).

The feasibility of copper bioleaching from fine milled PCB at low solids concentrations (up to 10.0 g/L) has been demonstrated and extractions as high as 90% were reported with mesophilic strains (Bas et al., 2013; Brandl et al., 2001; Xiang et al., 2010; Zhu et al., 2011). Nevertheless, quite few studies dealing with increased solid concentrations have been published so far likely because bacterial growth is inhibited in the presence of fine PCB particles. Although the exact nature of such deleterious effects is still unclear there are indications that are more important in the very beginning of

leaching experiments. This is because the quick release of harmful-to-bacteria species from the ground PCB is stimulated.

With the above mentioned concerns in the background, the current work proposes a different approach to PCB bioleaching, in which relatively coarse PCB sheets are leached without reducing their size down to the micrometer scale. The objective is to avoid useless overgrinding and hence reduce the overall cost associated with the fragmentation step. Another aim is to discuss the bacterial adaptation to increased PCB content and to find out the optimal pH and ferrous iron concentration for bioleaching using moderate thermophiles in a rotating-drum reactor.

2. Experimental procedure

2.1. PCB fragmentation and pre-treatment

The experiments were carried out with PCB collected from obsolete desktop computers, from which all electronic components such as capacitors and resistors were manually removed in advance. At the beginning, the dismantled boards were shredded using a metal guillotine to obtain nearly rectangular PCB fragments with 20 mm-long size. For the experiments with ground PCB, a fraction from the above-mentioned sheets was further fragmented using a laboratory hammer mill and after dry sieving the –208 μm + 147 μm particle size was selected. A second sample was subjected to a “pre-weakening” process in a laboratory jaw crusher (discharge gap 10 mm) aiming to generate cracks and to expose metals inside the PCB sheets to the leaching solutions. After jaw crusher “pre-weakening”, the samples were screened at 20 mm and the oversize fraction used for further studies. One part of this oversize material (fraction +20 mm) was subsequently submitted to a chemical pre-treatment step to remove the lacquer coating, which covered the printed circuit boards. PCB sheets (50 g) were mixed with 500 mL of aqueous solution of diethylene glycol (20% v/v) and potassium hydroxide (20% w/v) for accomplishing this task. The lacquer coating removal step was carried out under stirring at 90 °C during in 60 min. Thereafter the solid phase was filtered, washed with distilled water and then dried at 50 °C to a constant weight.

2.2. Microorganisms

The mesophile microorganisms utilized in the current study were isolated from a Brazilian sulfide mine, for which microbial

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