



The use of pyrite as a source of lixiviant in the bioleaching of electronic waste



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ABSTRACT

Electronic waste (e-waste) contains a wide range of elements, many of which are highly toxic to environmental and human health. On the other hand e-waste represents a significant potential source of valuable metals. This study used microbial oxidation of pyrite to generate a biolixiviant. Its efficiency in the dissolution of metals from printed circuit boards (PCBs) was evaluated as well as the effects of metals and PCB concentrations on microbial activity. The addition of elemental metals (Cu, Cr, Ni, Sn, Zn) had an immediate inhibitory effect on pyrite oxidation, though leaching recovered after a period of adaptation. Bioleaching was inhibited initially by the addition of 1 % (w/v) ground PCB, but recovered rapidly, whereas pulp densities of ≥ 5 % had sustained negative impacts on culture activity and viability. The loss of culture viability meant that only abiotic copper dissolution occurred at ≥ 5 % PCB. Final copper recoveries declined with increasing PCB pulp density. The relatively high content of elemental iron caused a lag period in copper solubilisation possibly due to displacement reactions. *Leptospirillum ferriphilum* was primarily responsible for pyrite oxidation, and most affected by both the pure metals (particularly Ni and Cu) and PCB.

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1. Introduction

Globally an estimated 35 million tonnes of electronic waste (e-waste) was produced in 2005 (Breivik et al., 2014), increasing to 49 million tonnes in 2012 (StEP, 2014). It makes up 1–3 % (8 % in rich countries) of municipal waste (Robinson, 2009; Widmer et al., 2005). Estimates vary on the amount of e-waste exported from developed nations rather than treated domestically. For example, available data from Eurostat¹ suggest that less than 1 % of e-waste is shipped outside of EU and approximately 58 % is recycled or reused. However, 13 % of collected e-waste is not reported as being processed following collection (within or outside the EU), so must presumably have been sent to landfill, stockpiled or otherwise ‘lost’. Breivik et al. (2014) suggest that 23 % of e-waste generated in developed nations is shipped to non-developed nations (principally China, India and several West African countries). Either way, such countries receive a disproportionate amount of the world’s e-waste. Lacking suitable facilities to treat it, it is invariably burnt, buried, dumped in water courses or processed in “backyard recycling facilities” which are “inefficient and polluting” (Nnorom and Osibanjo, 2008).

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¹ http://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=env_waselee (10th October, 2014).

E-waste has been reported as the largest emerging waste issue in Australia (Angel, 2008). The country produced 106,000 tonnes of e-waste between 2007 and 2008, but with limited domestic facilities recycled only around 10% (Khaliq et al., 2014; Li et al., 2013). With an average copper concentration of around 13% (after Cui and Zhang, 2008) and current copper prices around US\$6759.50²/tonne this means that over US\$103M copper was sent to landfill, incineration, storage or export in Australia alone. This presents a huge opportunity for valuable metal recovery.

E-waste comprises discarded large and small household appliances, IT and telecommunication equipment, consumer equipment, lighting equipment, electrical and electronic tools, toys, leisure and sports equipment, medical devices, monitoring and control instruments and automatic dispensers; or “anything with a battery or a cord” (Premalatha et al., 2013; StEP, 2014). Such items are made up of many individual components. These include printed circuit boards (PCBs), cathode ray tubes (CRTs; from older PC monitors and TVs), batteries, internal and external wiring and the equipment casing. Each has its own unique composition and associated environmental hazards (Cui and Zhang, 2008).

Many of the metals in e-waste are highly valuable and with available ore grades continuously decreasing there is a growing interest in recovering metal resources from e-waste. PCBs are a mixture of polymers, ceramics and metals tightly bonded together (Luda, 2011; Marques

² London Metal Exchange official settlement price (10th October, 2014).

et al., 2013). They make up just 3 % of e-waste mass, but account for the major fraction of valuable metals (Luda, 2011; Tuncuk et al., 2012; Vestola et al., 2010). Therefore, PCBs are most frequently targeted in recycling efforts.

Scrap PCBs are routinely divided into three categories based on their precious metal content (Kellner, 2008). High and medium grade materials have relatively high gold and palladium content (which make up 90 % of the inherent value of PCBs). In high grade scrap, this metal content is usually in discrete, readily separable components such as gold-containing integrated circuits, gold or palladium pin boards and thermally coupled modules from mainframes. Medium grade scrap has high precious metal content in pin and edge connectors with little incumbent material such as aluminium capacitors.

Most PCBs are sent to commercial smelters which typically credit between 92 % and 98 % of the precious metal value (Kellner, 2008). Low grade PCB scrap usually comes from televisions, other white goods and power supply units with heavy ferrite transformers and large aluminium heat sink assemblies. They have a low intrinsic value (i.e. relatively low precious metal content) which is insufficient to cover the smelting levies. Therefore, low grade PCB scrap is usually passed on at a loss by e-waste merchants.

Biohydrometallurgy uses microorganisms to generate a lixiviant which either causes the dissolution of the target metal or metals (for example copper from chalcocite) or (partially) liberates them from a matrix within which they are entrained (for example gold within arsenopyrite; Rossi, 1990). Since the 1992 Earth Summit in Rio, the concept of sustainable technologies and development has become very popular (Brombacher et al., 1997). Biohydrometallurgy finds favour in this respect as it is “almost without exception more environmentally friendly” than physicochemical processes (Rawlings, 2002). The process does not require the huge amounts of energy expended during roasting or smelting and does not produce harmful gas emissions. It can be done at standard temperature and pressure, does not require expensive or environmentally damaging reagents and traditionally offers lower operating expenditures as a result.

Studies into the application of biohydrometallurgy to e-waste have mainly involved the treatment of PCBs. Typically, the lixiviant is ferric iron from the biological oxidation of FeSO_4 and proton acidity through direct sulphuric acid addition or the biological oxidation of elemental sulphur. There is no clear advantage of using the microorganisms to generate ferric iron from ferrous sulphate, over the use of ferric sulphate directly (especially given the high degree of pH control required). Alternative and more sustainable sources of growth substrates and acidity are therefore desirable. Pyrite may provide a suitable source of growth substrate and acid, as microorganisms can oxidise the iron and sulphur in pyrite as an energy source and produce acidity, reducing the amount of pH control required.

While biohydrometallurgical recovery has been shown to be technically possible, many challenges remain; from substrate toxicity to downstream processing of the leach liquor. In their most simple form, the tests so far have involved liquid media containing ferrous iron in shake flasks, which may be amended with elemental sulphur and inoculated with pure or mixed cultures of iron- and sulphur-oxidising microorganisms. Ground PCB is either added immediately, in a one-step process, or following initial substrate oxidation (and thus lixiviant production) in a two-step or multi-step process.

In one-step tests, reported copper recovery efficiencies vary widely from less than 4 % to 100 % (Rivero-Hudec et al., 2009; Wang et al., 2009; Xiang et al., 2010; Zhu et al., 2011) with leaching times typically greater than 10 days and pulp densities of 1 % or less. Performance decreases significantly with increasing pulp density (Brandl et al., 2001; Liang et al., 2010; Wang et al., 2009; Xiang et al., 2010; Zhu et al., 2011). The waste tends to be acid-consuming and leaching may be better where constant pH is maintained (Yang et al., 2009), but this is not universal (Vestola et al., 2010). Several studies have demonstrated improved leaching performance in media containing elemental sulphur

as well as ferrous iron (Ilyas et al., 2007; Liang et al., 2010; Wang et al., 2009). Its inclusion in the medium has also been shown to reduce or even overcome the acid demand (Ilyas et al., 2007) and also provides an additional source of energy for organisms capable of sulphur oxidation. This may improve microbial resistance to various factors as many resistance mechanisms are metabolically active processes.

The toxicity of the PCBs to the microorganisms has been thought to be the major problem preventing efficient leaching, though it is not clear if this is due to the metallic or organic content. The metals tend to be in their elemental forms, making up on average 28 % (w/w; copper: 10–20 %, lead: 1–5 %, nickel: 1–3 %) of the mass (Ludwig et al., 2003). Ilyas et al. (2007) demonstrated improved bioleaching of crushed PCBs when the culture was adapted to high metal concentrations (up to 90 % copper recovery from a 10 % pulp density within 18 days). However, this included pre-washing the crushed PCBs in saturated NaCl which removed a large fraction of the organic content. Moreover, the PCBs contained relatively low levels of copper (8.5 % w/w). The data suggested that improved bioleaching was due to both culture adaptation to the metals and proportional reduction of some of the non-metallic fraction.

Staggering the production of the lixiviant and the addition of the e-waste in a two-step process has been shown to greatly increase final copper recoveries and leaching rates (Liang et al., 2010; Xiang et al., 2010; Yang et al., 2009; Zhu et al., 2011) but this has not been tested at pulp densities greater than 2 %. Furthermore, if the addition of the e-waste at the second step results in a loss of culture viability, then fresh media and inocula will be required for each subsequent run and it may not be possible to subculture from one run to the next.

This study evaluates the use of pyrite as the source of lixiviant in the bioleaching of low-grade electronic wastes which cannot be sold profitably by e-waste merchants. The effects of various key metals on pyrite dissolution and microbial activity were assessed and PCB leaching rates, acid consumption and final metal recoveries determined as a function of sulphide substrate and pulp density. Finally, the effects of PCB and pure metal leaching on the microbial community were examined.

2. Materials and methods

2.1. Electronic waste and sulphide minerals

Shredded, low-grade, waste PCBs were collected from Total Green Recycling, Perth, Western Australia and the <10 mm fraction milled in a ball mill for 2 h. The resulting <2 mm fraction was used in bioleaching experiments. Size distribution and total metal content (dry ashing followed by aqua regia digestion) were determined by CSIRO Process Science and Engineering, Perth, Australia and are shown in Tables 1 and 2. The low copper content is typical of low-grade e-waste fractions. Pyrite concentrate (60 % pyrite with 12.5 % quartz, 9 % albite and 7.5 % dolomite the other major minerals; p80 passing 120 μm) was sourced from Kalgoorlie Consolidated Gold Mines Pty Ltd (KCGM), Australia.

2.2. Microbial culture

Stock cultures used to inoculate the bioleaching tests were maintained in BSM medium ($(\text{NH}_4)_2\text{SO}_4$ [1.5 g L^{-1}], $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ [0.25 g L^{-1}], KH_2PO_4 [0.25 g L^{-1}]; Keeling et al., 2006), pH 1.7, containing 3 % w/v pyrite with 0.1 % w/v ground PCB, under non-sterile conditions. The original culture contained *Acidithiobacillus* (*At.*) *calculus* BRGM3, *Leptospirillum* (*L.*) *ferriphilum* BRGM1, *Sulfobacillus* (*Sb.*) *benefaciens* BRGM2 and *Ferroplasma* (*Fp.*) *acidiphilum* BRGM4 (a stock consortium isolated from a commercial bioleaching reactor treating cobaltiferous pyrite, which had been exposed to e-waste in the form of shredded cables from automotive recycling for over a year), *At. ferrooxidans*^T DSM14882 (due to its mesophilic growth range and ability to thrive in low redox conditions; Rawlings et al., 1999) and *Acidiphilium* sp. SJH (due to its ability to break down organic compounds which may inhibit the activity

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