



## New approaches for extracting and recovering metals from mine tailings



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### ABSTRACT

Waste materials from metal mining, such as mineral tailings, often contain significant amounts of potentially valuable metals particularly where, as in many historic operations, the efficiency of flotation technologies used to concentrate target minerals was not as good as those currently available. A two-stage mineral leaching and metal recovery protocol was developed to extract copper from tailings generated as waste materials in two mines currently operating in Spain and Serbia. The most effective extraction of copper (84 to >90%) was achieved by bioleaching the tailings at 45 °C, using a defined microbial consortium, where elemental sulfur was added to the tailings and the pH of leach liquors allowed to fall to ~pH 1, at which point anaerobic conditions were imposed. The thermo-tolerant acidophiles *Acidithiobacillus caldus* and *Sulfobacillus thermosulfidooxidans* emerged as the dominant bacteria present in both tailings leachates under these conditions. Copper present in the pregnant leach solutions (PLS) produced were next precipitated as a sulfide phase using hydrogen sulfide generated in a low pH (4.0) sulfidogenic bioreactor. The off-line system used allowed the copper present in PLS to be precipitated selectively without the need to adjust the pH of the PLS, though small amounts of silver present in PLS from one of the tailings samples co-precipitated with copper sulfide. Experimental data also suggested that it would be possible to extract silver from bioleached solid residues (where it was mostly found) using a simple chemical extractant. The results suggested that bio-processing these waste materials would have economic as well as environmental benefits.

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### 1. Introduction

Mining of metals generates considerably amounts of waste materials. These generally have very little economic value, making their exploitation not profitable, though they often have the potential to pose a long-term threat and cause damage to the environment. Mine wastes vary depending on their physical and chemical composition, the type of mining and the way the mineral is processed. Millions of tonnes of ore are processed every year by the mining industry, >95% of which is disposed of in the form of waste rocks and mine tailings. The latter are finely ground rock particles generated during the processing of ore materials and separation of target metal minerals, and are highly reactive because of their small particle size and content of reactive minerals, such as pyrite (FeS<sub>2</sub>). Mine tailings may contain base transition metals, such as iron, copper, nickel and zinc, in relatively high concentrations, and also occasionally precious metals such as gold and silver, in minerals (and native metals) that have been not been separated

by froth flotation (e.g. Bryan et al., 2006; Ahmadi et al., 2015). Toxic elements, such as arsenic, may also be present in elevated concentrations (e.g. Nguyen et al., 2015). Exposure of mine tailings to both oxygen and water facilitates the (microbially-catalysed) oxidative dissolution of the sulfide minerals they contain, generating acidic waste waters that contain elevated concentrations of dissolved metals and sulfate. Acid mine drainage (AMD) is a widely-reported environmental pollutant, with global distribution.

Storage of waste rocks and mineral tailings at, and in the vicinity of, mine sites represents a long-term engineering and environmental challenge. Occasionally, catastrophic environmental pollution has occurred due to the failings of the system used to store mine wastes. One such example is the failure of the retaining dam of a tailings impoundment at the Aznalcóllar-Los Frailes mine (in south-west Spain) which resulted in the release of vast amounts of acidic waters and tailings slurries into the catchment of a local major river nearby the Doñana National Park in April 1998 (Grimalt et al., 1999; Ericksson and Adamek, 2000). However, mine tailings may also be recycled and reused, e.g. manganese-rich tailings could be used in agro-forestry, building and construction materials, coatings, cast resin products, glass, ceramics, and glazes (Lottermoser, 2011). Increasing demands for metals, coupled with

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depleting reserves of high-grade primary ores, has meant that waste materials from historic mining operations may have greater metal contents than primary ores currently being excavated. Since mine tailings have already been part-processed (by comminution, to produce fine-grain particles) the cost of extracting residual metals from them is often economically more attractive than mining a deep-buried primary ore body. In addition, mine wastes may contain metals (such as rare earth elements) that were not considered worth extracting when the ores were initially processed, but which have since increased in value and use. This was the case with cobalt-rich tailings produced at a former copper mine in Kasese, Uganda, which were reprocessed by bioleaching decades after mining at the site ended, both to extract cobalt and to remove the environmental threat posed by the tailings deposits (Morin and D'Hugues, 2007).

Harnessing the abilities of some species of prokaryotic microorganisms to catalyse the oxidative dissolution of sulfide minerals and thereby to facilitate the extraction of metals is a global biotechnology, used mostly to recover some base including copper, nickel, cobalt and zinc, and precious metals such as gold. The geochemical processes and the microorganisms involved in biomining have been widely researched and are well understood (e.g. Vera et al., 2013; Johnson, 2014). However, not all sulfide minerals are readily bioleached such as chalcopyrite. This mineral (the most abundant copper mineral) is notoriously difficult to effectively oxidise using conventional bioleaching approaches (reviewed in Watling, 2006). Recently, extraction of metals from oxidised ores using reductive, rather than oxidative, bioleaching has been described (Johnson et al., 2013). In contrast to conventional biomining, this requires the addition of an extraneous material, such as elemental sulfur, to act as the electron donor for bacteria that reduce ferric iron minerals.

In this work, we describe results from bioleaching mine tailings generated at two operational copper mines, carried out at different temperatures (with mesophilic and moderately thermophilic acidophilic microorganisms) and using a combination of oxidative and reductive bioleaching.

## 2. Materials and methods

### 2.1. Mineral tailings

Mineral tailings were obtained from two copper mines, Bor (Serbia; operated by RTB Bor), and Cobre Las Cruces (CLC; Sevilla, Spain; operated by First Quantum Minerals Ltd.).

### 2.2. Bioleaching microbial consortia

Three different consortia of acidophilic microorganisms were used in bioleaching experiments. These were: (i) a thermo-tolerant consortium, which included the autotrophic iron-oxidizer *Leptospirillum* (*L.*) *ferriphilum* (both the type strain and strain MT63), the autotrophic sulfur-oxidizer *Acidithiobacillus* (*At.*) *caldus*<sup>T</sup>, facultatively autotrophic and obligately heterotrophic iron/sulfur oxidizing and iron-reducing bacteria (*Sulfobacillus* (*Sb.*) *thermosulfidooxidans*<sup>T</sup> and *Acidibacillus* (*Ab.*) *sulfuroxidans*<sup>T</sup>, and two heterotrophic iron-oxidizing archaea: *Ferroplasma* (*F.*) *acidiphilum* (strain BRGM4) and *Acidiplasma* sp. (strain FV); (ii) a second thermo-tolerant consortium, used in bioreactors where pH was not controlled, that included *L. ferriphilum* strain MT63, *At. caldus*<sup>T</sup>, *Sb. thermosulfidooxidans*<sup>T</sup>, “*Ab. sulfuroxidans*”<sup>T</sup>, *F. acidiphilum* strain BRGM4, *Acidiplasma* strain FV, and two strains of *Sulfobacillus* that had been isolated from the Bor tailings (*Sb. thermosulfidooxidans* strain BOR3 and *Sb. acidophilus* strain BOR1); (iii) a mixed mesophilic/thermo-tolerant consortium, that included six species

of mesophilic acidophiles (autotrophic iron/sulfur-oxidizing and iron-reducing *Acidithiobacillus* spp. (*At. ferrooxidans*<sup>T</sup>, *At. ferridurans*<sup>T</sup>, *At. ferriphilus*<sup>T</sup> and *At. ferrivorans* strain CF27), “*Ab. ferrooxidans*”<sup>T</sup> (a heterotrophic iron-oxidizer/reducer), *Acidiphilum* (*A.*) *cryptum* (strain SJH; a heterotrophic iron-reducer), and three thermo-tolerant species (*Sb. thermosulfidooxidans*<sup>T</sup>, *F. acidiphilum* strain BRGM4 and *Acidiplasma* strain FV). The consortia were grown in iron/yeast extract media pH 2.0, at 30 (mesophilic/thermo-tolerant consortium) or 45 °C (thermo-tolerant consortia) for 2–3 weeks previously to inoculation of the reactors.

### 2.3. Bioleaching experiments and processing of mineral residues

Bioleaching of mineral tailings was carried out in 2.3 L bioreactor vessels coupled to temperature- and pH-control units (Electrolab, UK). Temperatures were maintained at either 30° (for the mesophilic/thermo-tolerant consortium) or 45 °C (for the thermo-tolerant consortia), and the reactors were stirred continuously at 150 rpm. Control of pH (where used) was by automated addition of 0.5 M sulfuric acid or 0.5 M sodium hydroxide. For aerobic conditions, the reactor vessels were gassed with sterile air, and when anoxic conditions were required this was replaced by oxygen-free nitrogen (OFN), both at flow rates of ~1 L/min. A basal salts/trace elements solution (Nancuqueo et al., 2016) was put into each reactor vessel (1.4 L for aerobic bioleaching, and 1.9 L for combined aerobic/anaerobic bioleaching), followed by mineral tailings (5% pulp densities; w/v) and 100 mL of pre-grown microbial inocula.

Six bioleaching experiments were carried out in total, three with Bor tailings and three with CLC tailings. These were: (i) aerobic bioleaching, carried out at 30 °C and pH 1.7; (ii) aerobic bioleaching, carried out at 45 °C and pH 1.7; (iii) alternated aerobic/anaerobic bioleaching, carried out at 45 °C with an initial pH of 1.8 but with pH control removed and elemental sulfur added to the tailings (at 2%, w/v) to act as an electron donor under both aerobic (for acid production) and anaerobic (for ferric iron reduction) conditions. In the last set-up, the bioreactors were aerated up to the point until the pH had declined to 1.0, at which point the air was replaced with OFN. Samples were withdrawn from the bioreactors at regular intervals to measure redox potentials and pH values (off-line), concentrations of soluble transition metals (iron, copper, zinc, manganese), and also iron speciation.

When the bioleaching phase was considered to be complete, mineral slurries were removed from the bioreactor vessels, and the pregnant leach solutions (PLS) separated from the mineral residues. Tests were carried out to selectively precipitate copper from PLS by sulfide precipitation (Section 2.4). Copper and silver were extracted both from mineral residues and fresh mineral tailings by mixing 0.5 g of representative samples with 10 mL of a solution containing 3.42 M NaCl and 0.2 M HCl (Romero et al., 2003), and heating for 1 h at 90 °C.

### 2.4. Sulfidogenic bioreactors and selective recovery of copper from PLS

Low pH, continuous-flow sulfidogenic bioreactors (Fig. 1) were used as sources of hydrogen sulfide (H<sub>2</sub>S) for precipitating copper present in PLS generated in the bioleaching experiments described above. Details of the design and operation of these biosulfidogenic reactors are given elsewhere (e.g. Nancuqueo and Johnson, 2012). In brief, these were 2.2 L (working volume) bioreactors coupled to FerMac 310/60 control units (Electrolab, UK), maintained at pH 4.0 and fed with an acidic (pH ~ 2.5) liquid medium containing 5 mM glycerol (the electron donor used by the sulfidogenic bacteria in the bioreactor). The H<sub>2</sub>S generated in the reactor vessels was transported in a continuous flow of OFN into off-line gas bottles that contained samples of PLS samples.

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