



Acid leaching of a copper ore by sulphur-oxidizing microorganisms



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ARTICLE INFO

Article history:

Received 18 February 2014
Received in revised form 21 May 2014
Accepted 28 May 2014
Available online 14 June 2014

Keywords:

Bioleaching
Sulphuric acid leaching
Biooxidation of sulphur
Copper bioleaching

ABSTRACT

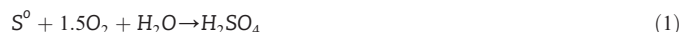
This study examines the potential for use of biogenic sulphuric acid for leaching of a low-grade Brazilian ore containing 0.64% copper occurring as brochantite, $\text{Cu}_4\text{SO}_4(\text{OH})_6$, and as malachite, $\text{Cu}_2\text{CO}_3(\text{OH})_2$. Ore was agglomerated with elemental sulphur and sulphur-oxidizing microorganisms. Copper extraction was more efficient from inoculated ore amended with 6.67 kg S^0 /t of ore, compared to uninoculated ore with no added S^0 and leached with an acid solution maintained at pH 2.0 with sulphuric acid. The rate and extent of copper bioleaching were proportional to the amount of added S^0 . A maximum of 88% copper was extracted after 9 weeks of bioleaching with 13.3 kg S added/t of ore. Cu extraction was more efficient if sulphur was uniformly mixed with the ore than if the sulphur was added only to the upper zone of the ore column. There was no evidence that acid leaching of Cu from this ore was far more efficient in ore crushed by a high pressure grinding rolls (HPGR) compared to a conventional jaw crusher.

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1. Introduction

Biooxidation of metal sulphide ores and concentrates is practiced at industrial scale in stirred tank reactors and in heaps (Brierley and Brierley, 2013). The microbiological contribution to metal sulphide oxidation is largely a result of the elevated solution redox potential produced via biooxidation of ferrous iron (generally derived mainly from pyrite) to ferric iron at low pH.

Conversely, much less work has been done on leaching of acid-soluble metals from ores by biogenic sulphuric acid. The acid is most efficiently produced biologically via oxidation of elemental sulphur by *Acidithiobacillus thiooxidans* and *At. caldus*, which grow optimally at about 30 °C and 45 °C, respectively. These bacteria use elemental sulphur as an energy source, oxidizing it to sulphate under aerobic conditions with oxygen as the electron acceptor (reaction 1).



This reaction produces 530 kJ of heat per mole of S oxidized to sulphate and one tonne of sulphuric acid per 327 kg of S^0 oxidized (West-Sells et al., 2007).

Metals from contaminated soils, sediments or sewage sludge can be leached by biooxidation of S^0 added to the solids (Blais et al., 1992; Seidel et al., 2006; Tichy et al., 1998a). Recovery of metals from highly acid-consuming sulphide ores also benefits from microbial acid

production. Biooxidation of added S^0 overcame the acid demand of a low-grade, sulphidic Ni ore with high acid demand (Salo-Zieman et al., 2006). This approach represents a potential bioheap leaching method of Ni recovery from this ore. Under certain conditions, addition of S^0 to a chalcopyrite concentrate enhances the leaching rate, attributed in part to reduced Fe^{3+} precipitation (Vilcaez et al., 2008). The heat produced from adding S^0 to a heap of base metal sulphide ore may improve bioleaching rates (Williams, 2004).

Microbial production of sulphuric acid is relevant to the mining industry as sulphuric acid is used extensively, particularly in heap and dump leaching of copper. The Chilean mining industry is the world's most intensive sulphuric acid user (Chilean Copper Commission, 2009). Sulphuric acid is amongst the largest operating costs in copper leach operations (West-Sells et al., 2007). However, economic comparisons between purchased sulphuric acid and sulphuric acid produced from S^0 biooxidation are not straightforward. The costs of these commodities vary considerably from year to year and with distance from smelters. And, of course, sulphuric acid streams produced biologically are dilute and take time to produce. Nonetheless, assuming the costs per tonne of S^0 and H_2SO_4 are similar, sulphuric acid production from S^0 is 67% cheaper than sulphuric acid (reaction 1, West-Sells et al., 2007).

Microorganisms may also be used to biooxidize S^0 in reactors to produce a “manufactured” stream of dilute sulphuric acid for leaching purposes. Several investigations have aimed to improve the relatively slow kinetics of microbial S^0 oxidation in stirred reactors (Gourdon and Funtowicz, 1998; Liu et al., 2004; Tichy et al., 1998b). Biooxidation of S^0 by *At. caldus* in an aerated batch reactor produced an average sulphuric acid concentration of 0.45 M. This solution was used to leach

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the $^7\text{Li}^+$ isotope from degraded resin (Basitere et al., 2012). Phelps Dodge Mining Company developed a stirred and aerated reactor for microbial sulphuric acid production. Solids (S^0) were mechanically retained in the production vessel; this decoupling of solids and liquid retention times improved rates of microbial sulphur conversion to sulphuric acid (Uhrrie, 2004; Uhrrie et al., 2004). Acid concentration was kept preferably to less than 35 g/L so as not to exceed microbial acid tolerance levels. Biooxidation of S^0 or other reduced sulphur compounds in aerated piles, heaps or trickle bed reactors might also be used to produce sulphuric acid for leaching purposes (Kelso and Parkison, 2008; Young et al., 2003a,b).

As compared to application of sulphuric acid solutions to heaps, the localized generation of sulphuric acid from S^0 biooxidation within heaps of ore containing acid-soluble copper has several potential advantages: 1) sulphuric acid applied only to the top of a heap is depleted as leach solutions move downward through copper ore heaps (Bartlett, 1995), 2) addition of large quantities of liquid sulphuric acid to a heap may be compromise heap stability (Salo-Zieman et al., 2006), 3) heat is produced from biooxidation of S^0 (West-Sells et al., 2007; Williams, 2004), increasing reaction kinetics, 4) acid consumption will be less if sulphuric acid is generated and consumed locally at reactive mineral sites, thereby maintaining low acid concentrations and a low driving force for acid/gangue reactions (Bruynesteyn, 2004), 5) transportation, storage and use of solid S^0 are safer than hazardous H_2SO_4 (Salo-Zieman et al., 2006).

The in situ biological production of sulphuric acid may begin with conditioning of S^0 with a bacterial solution to facilitate attachment of microorganisms and wetting of the hydrophobic sulphur particles, followed by agglomeration with the ore (Bruynesteyn, 2004). Using this approach, column tests with Barrick's Zaldivar copper ore (0.68% Cu) showed acid could be produced by biooxidation of added S^0 whether sulphur was replacing sulphuric acid or added in excess (West-Sells et al., 2007). Up to 75% of the sulphuric acid normally added to agglomeration was substituted with S^0 and did not appear to inhibit Cu leaching even in columns as tall as 5.1 m. Columns were aerated and held at 25 °C. Copper in this ore was 27% soluble in acid and 62% soluble in cyanide (assumed to be chalcocite or covellite).

Other than this Zaldivar work, little has been published showing the extent that acid-soluble copper can be leached from ores agglomerated with S^0 and bacteria to produce sulphuric acid. This paper describes column tests with the addition of sulphur and sulphur-oxidizing bacteria to a Brazilian copper ore in which copper is mostly acid-soluble. Copper leaching with biogenic sulphuric acid at various S^0 loadings is compared to leaching only with reagent sulphuric acid added to leach solutions.

Additionally, we tested whether grinding of ore with high pressure grinding rolls (HPGR) could improve copper extraction from leaching with biologically produced acid. HPGR introduces micro-fractures in the ore particles, improving access of leach solutions to the interior of the particles. However, the HPGR product can produce more fines during the leaching process that could have adverse effects on heap percolation (Ghorbani et al., 2013). Nonetheless, the technology is becoming widely adopted in ore processing operations and applied to both base metal (Baum and Ausburn, 2011) and precious metal (van der Meer and Maphosa, 2011) operations.

2. Experimental

2.1. Organisms and culture conditions

Two microbial cultures were used in the tests. One was a pure culture of *At. thiooxidans* (FG01) initially isolated from an acid effluent from a uranium mine (Garcia, 1991). It was maintained in shake flasks at 30 °C on modified Kelly medium (MKM) containing elemental sulphur as an energy source (1% w/v). MKM contains 0.4 g/L ammonium sulphate, 0.4 g/L magnesium sulphate heptahydrate and 0.04 g/L potassium dihydrogen phosphate. The pH of MKM was adjusted to 2.0 with 5 M sulphuric acid solution.

A mixed culture of mesophilic metal sulphide-oxidizing bacteria was also utilized. The culture originally contained *At. thiooxidans* and *At. ferrooxidans* along with species of iron-oxidizing *Leptospirillum* and *Ferropasma*. The mixed culture had been maintained on MKM with ferrous iron, sulphur and pyrite. It was transferred to MKM plus S^0 and 100 mg/L dissolved Cu (as copper sulphate pentahydrate) and grown for 3 days prior to ore leaching tests.

Bacterial cell numbers in solutions were determined by phase contrast microscopy using a Thoma counting cell.

2.2. Ore

A 200 kg sample of bulk copper ore was received from the Caraiba Surubim Mine in Bahia State, Brazil in a particle size ranging from 25.4 to 38.1 mm (1.0 to 1.5 in). This sample was thoroughly mixed and sub-samples were removed for analysis (after crushing and pulverizing) and for column tests.

The ore sample contained 0.64% Cu (Table 1) as determined by atomic absorption analysis following acid-digestion of a blended sample (Taylor and Reisman, 2000). Copper occurred as brochantite $\text{Cu}_4\text{SO}_4(\text{OH})_6$ and as malachite $\text{Cu}_2\text{CO}_3(\text{OH})_2$ as determined by X-ray diffraction (XRD). Although XRD detected no other copper minerals, a small amount of copper (2.5% of the Cu in the ore) was cyanide soluble after sulphuric acid leaching. The ore also contained 2.20% S (determined with a Leco SC632 sulphur analyser). Other metals were determined by X-ray fluorescence.

Subsamples of the bulk ore removed for column tests were crushed with a jaw crusher or with high pressure grinding rolls (HPGR). The crushed ore was screened to a particle size of 12.7 mm \times 3.36 mm. Sub-samples of this size fraction were removed for copper analysis and confirmed the Cu head analysis of the bulk sample. Fines were eliminated to more easily discern the effects of acid leaching of Cu from a discrete ore particle size range. The acid consumption of the ore (jaw crushed) was determined by adding 0.5 M sulphuric acid to a slurry of pulverized ore (40 g) in water (250 mL) in a 500 mL beaker until a stable pH of 2.0 was achieved.

2.3. Conditioning of sulphur

Elemental sulphur was added (10 to 40 g, depending on the experiment) to 30 mL of an active microbial culture growing on MKM-S (10^9 cells/mL) plus 70 mL MKM salts (pH 2.0) in a 250 mL Erlenmeyer flask. The slurry was incubated at 30 °C with shaking at 150 rpm for 18 to 24 h, thereby conditioning (inoculating and wetting) the sulphur (Bruynesteyn, 2004).

2.4. Agglomeration

Ore (3.0 kg) was agglomerated with 100 mL of the conditioned sulphur slurry by slowly adding the slurry to ore in a rotary mixing chamber. Our initial tests showed that this ratio of solution to ore was just enough to produce a moist ore charge coated with sulphur without free moisture left over. Starting bacterial density in inoculated columns was approximately 10^7 cells/g ore. Control columns were uninoculated and did not receive sulphur. The ore in these columns was either agglomerated with pH 2.0 (sulphuric acid) water or not agglomerated with a deionized water leach.

Table 1
Analysis of Surubim Mine copper ore.

% Cu	% S	% Fe	% Ca	% Mg	% K
0.64	2.20	4.57	0.50	2.57	0.88

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