



Silver-catalyzed bioleaching of copper, molybdenum and rhenium from a chalcopyrite–molybdenite concentrate



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ABSTRACT

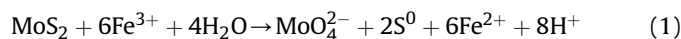
This study evaluated silver-catalyzed bioleaching to solubilize copper from a chalcopyrite-bearing molybdenite concentrate with mesophilic (32 °C) and moderately thermophilic (45 °C) microbial consortia. The Cu content of the concentrate was 0.98% (wt/wt), with chalcopyrite as the major Cu-bearing mineral. The molybdenite phase (53.8% Mo) also contained 0.055% rhenium. The addition of Ag⁺ as silver nitrate greatly enhanced copper dissolution under bioleaching conditions. Overall, >93% Cu was leached in the bioleaching experiments involving the addition of up to 200 mg l⁻¹ Ag, contrasted by 53% Cu solubilization in the absence of silver. The highest relative yields (>99%) were obtained under moderately thermophilic conditions. Different pulp densities (3–9%) and initial pH values (1.5–1.9) were also tested in the bioleaching experiments but their effects were masked by enhancement of copper leaching due to silver catalysis. Molybdenum and rhenium responded comparably to changes in bioleaching conditions, suggesting that Re was located in the molybdenite matrix. Their relative dissolution was <1% Mo and <6% Re, and only a minor enhancement of their dissolution was observed upon Ag addition. Compared with the bioleaching with mesophiles at 32 °C, the dissolution of Mo and Re was lower in the 45 °C experiments with moderate thermophiles. Because the corresponding yields of Cu dissolution increased with the temperature, this difference suggested early passivation of molybdenite, Mo precipitation, or a suppression of molybdenite oxidation capacity of the moderately thermophilic culture at the higher temperature.

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1. Introduction

Cu- and Fe-sulfides are typical impurities in molybdenite (MoS₂) concentrates and are difficult to remove adequately by conventional physical and chemical separation techniques. In principle, Cu- and Fe-sulfide minerals can be oxidized in bioleaching processes but concurrent solubilization of molybdenite may also occur. The leaching of mixed sulfide minerals involves galvanic interactions. In the electrochemical series of semiconductive sulfide minerals, molybdenite has a higher electrode potential than chalcopyrite (CuFeS₂). If only controlled by galvanic coupling,

molybdenite in contact with chalcopyrite acts as a cathode and is protected from oxidative leaching while chalcopyrite is anodically dissolved. However, many other factors interplay in the bioleaching process. The bioleaching of sulfide minerals involves oxidants such as Fe³⁺ and iron- and sulfur-oxidizing, acidophilic prokaryotes. Bioleaching of molybdenite in acid solutions leads to the formation of the molybdate ion (Romano et al., 2001a,b,c).



The bioleaching of molybdenite is mediated by ferric iron. The role of acidophilic prokaryotes in this process is to oxidize ferrous iron and sulfur compounds and help maintain high redox potential in favor of molybdenite oxidation as determined by the Fe³⁺/Fe²⁺ ratio. Mesophilic bioleaching of molybdenite was tested already in the mid-1950's with undefined iron- and pyrite-

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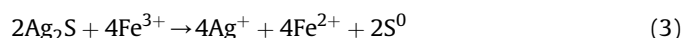
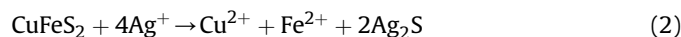
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oxidizing bacterial cultures (Bryner and Anderson, 1957; Bhappu et al., 1965). Brierley (1974) published results for the bioleaching of a molybdenite sample with a thermophilic archaeon, an *Acidianus* sp., tested at 60 °C. In general, molybdenite dissolution in bacterial or archaeal cultures was clearly higher than in abiotic controls. Molybdenite is considered to be relatively recalcitrant in bioleaching systems (Lasheen et al., 2015) and the reported yields and rates have not been sufficient to warrant further development for application.

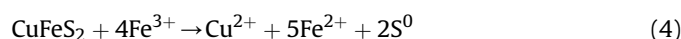
Molybdenite can be the host for rhenium, and minor amounts of Re are solubilized during the bioleaching process (Askari Zamani et al., 2005). Rhenium is associated with molybdenum as an isomorphous solid solution of ReS₂ in MoS₂ (Rouschias, 1974). Rhenium may also partially substitute for isovalent Mo in molybdenite (Voudouris et al., 2009; da Silva et al., 2013).

Selective bioleaching of the Cu-sulfide fraction is one of the potential pretreatments for upgrading Cu-containing molybdenite concentrates. Chalcopyrite associated with host molybdenite is a problem in this regard because of its refractory properties. The bioleaching of chalcopyrite is a relatively slow process compared to secondary Cu-sulfides. Secondary solid phases formed on the chalcopyrite surface contribute to the refractory properties of chalcopyrite. These secondary phases include the formation of elemental S and Fe-deficient, Cu-rich intermediate sulfides on chalcopyrite surface (Debernardi and Carlesi, 2013; Li et al., 2013; Yang et al., 2015). Secondary Cu-sulfides alter the semiconductive properties of the mineral surface and contribute to surface passivation. The formation of jarosite type precipitates on the chalcopyrite surfaces is also believed to contribute to passivation.

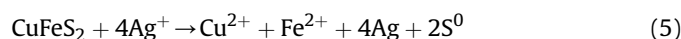
Elevated temperature and catalytic ions have been tested in attempts to alleviate the passivation effect on chalcopyrite surfaces. The use of thermoacidophilic iron- and sulfur-oxidizing archaea at high temperatures (Plumb et al., 2002, 2008; Norris et al., 2013) increases the rates of bioleaching and attenuates the passivation of chalcopyrite surfaces. Among catalytic ions, silver is of special interest because it has been proven effective and selective in enhancing Cu dissolution through the biological oxidation of chalcopyrite (Li et al., 2013). The positive catalytic effect of silver ions on the bioleaching of chalcopyrite has been demonstrated in numerous bioleaching experiments (e.g., Muñoz et al., 1998, 2007a,b; Córdoba et al., 2009; Feng et al., 2013). The mechanism of silver catalysis, as proposed by Miller et al. (1981) and Miller and Portillo (1981), is based on the reaction of Ag⁺ to form Ag-sulfide on the chalcopyrite surface, followed by Fe³⁺-mediated oxidation:



Silver recirculates between the solution and solid phase reactions and the overall sum of the chalcopyrite reaction yields elemental S:



Elemental Ag can also form during the process (Muñoz et al., 1979).



The concentration of dissolved silver is affected by the mineralogical composition of the ore and the formation of precipitates during the bioleaching. Ferric iron in the leach solution is subject to hydrolysis, leading to jarosite type Fe(III)-hydroxysulfates in sulfate-rich solutions in the presence of monovalent cations. Silver

can precipitate as argentojarosite with an ideal formula of AgFe₃(-SO₄)₂(OH)₆ in the bioleaching environment (Ahonen and Tuovinen, 1990; Sasaki et al., 1995; Córdoba et al., 2009) and may contribute to its depletion from the solution phase. Ag-jarosite lacks the catalytic effect on chalcopyrite leaching.

Our previous study demonstrated that silver addition enhances the bioleaching of Cu from a chalcopyrite-containing molybdenite concentrate (Abdollahi et al., 2014). The present work expands the previous study by examining the effect of several independent variables, most importantly the concentration of silver, initial pH, pulp density, and mesophilic and moderately thermophilic microbial consortia on the bioleaching of chalcopyrite. This study is part of a research program that examines the feasibility of using the silver catalysis combined with bioleaching technology as a pretreatment to selectively remove Cu-sulfides from molybdenite concentrates. While positive silver catalysis in the bioleaching of chalcopyrite has been demonstrated in numerous publications, much less is known of its potential effects on catalysis of Mo and Re dissolution, which were the other main metals of interest in the Cu-containing concentrate. A range of Ag concentration was tested, up to 1000 mg Ag l⁻¹, in an effort to characterize the catalyst effect on the bioleaching of Cu, Mo, and Re.

2. Materials and methods

2.1. Molybdenite concentrate

The sample of a chalcopyrite-containing molybdenite concentrate was provided by the Sarcheshmeh Copper Complex of Iran. The particle size distribution was 90% –38 μm. The concentrate sample contained (wt/wt) 53.8% Mo, 1.56% Fe, 0.98% Cu, and 0.055% Re. Molybdenite was the major phase (89.8% of the mineralogical composition), and minor phases were chalcopyrite (2.1%), pyrite (2.0%), and covellite (0.3%). As reported by Aminsazadeh et al. (2011) for the Sarcheshmeh Cu–Mo deposit, rhenium was associated with the molybdenite matrix but its further mineralogical analysis was not conducted. Optical microscopic examination indicated that the Cu- and Fe-sulfide minerals were mostly liberated from the molybdenite phase in this size fraction. These observations were also verified with EDAX-analyses, which were carried out with scanning electron microscopy and backscattered electron emission. The same bulk sample was used in our previous study of silver catalysis (Abdollahi et al., 2014).

2.2. Microorganisms

Two mixed cultures of acidophilic microorganisms, detailed by Abdollahi et al. (2014), were used in the bioleaching experiments. The mesophilic consortium originated from mine drainage of the Sarcheshmeh copper mine, Kerman Province. The consortium contained *Acidithiobacillus ferrooxidans*, *Acidithiobacillus thiooxidans* and *Leptospirillum ferrooxidans*. The consortium was cultured at 32 °C. The moderately thermophilic consortium was obtained from Mintek SA, Johannesburg, South Africa and it contained *Acidithiobacillus caldus*, *Leptospirillum ferriphilum*, and *Sulfobacillus* and *Ferroplasma* spp. This consortium was cultured at 45 °C. The cultures were maintained with 10% chalcopyrite concentrate as an energy source in shake flasks at 150 rpm. The mesophilic consortium was cultured in 9 K mineral salts at pH 1.8 (per liter: 3.0 g (NH₄)₂SO₄, 0.5 g MgSO₄·7H₂O, 0.1 g KCl, 0.5 g K₂HPO₄, and 0.01 g Ca(NO₃)₂). The moderately thermophilic consortium was maintained with chalcopyrite in mineral salts medium that contained (per liter) 0.2 g each of (NH₄)₂SO₄, MgSO₄·7H₂O, and K₂HPO₄ at pH 1.5. Both mixed cultures were gradually adapted to grow with up to 10%

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