



Microbial assisted galvanic leaching of chalcopyrite concentrate in continuously stirred bioreactors



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ABSTRACT

The objective of this study is to combine two proven technologies, namely galvanic and microbial assisted leaching of chalcopyrite in a novel way. Using a commercial chalcopyrite-rich flotation concentrate and pure natural pyrite samples from an Iranian copper mining complex as well as mixed cultures of moderately thermophilic microorganisms, continuous electrochemical bioleaching tests were performed in stirred bioreactors at 50 °C as a first step towards a commercial application of microbially assisted galvanic leaching of chalcopyrite.

From the results of this study, the importance of oxidation reduction potential (ORP) on the catalytic interaction between chalcopyrite and pyrite could be pointed out as a main parameter for successful bioprocessing of chalcopyrite concentrates. Optimization of the particle size of feed (D_{80}) and adjusting the ORP in the range between 400 and 500 mV are important criteria, governing the electrochemical bioleaching rate of chalcopyrite concentrates. It is believed that the main reason for increased copper recovery could be the control of chalcopyrite passivation resulting from improved galvanic interaction between chalcopyrite and pyrite in the selected ORP range and the bacterial assisted oxidation of Fe^{2+} to Fe^{3+} . Under optimum conditions of this study ($T = 50 \pm 2$ °C, $pH = 1.1 \pm 0.2$, $ORP = 400\text{--}500$ mV, solid load = 10%, stirring rate = 300 rpm) the copper extraction from chalcopyrite flotation concentrate during continuous electrochemical bioleaching operations in stirred bioreactors was > 95%, which should be high enough to justify the process economically.

1. Introduction

Copper is known as the third most worldwide used metal in terms of tonnage. It is usually present in the earth's crust as sulfide minerals mostly as chalcopyrite, which is usually beneficiated by flotation method to achieve a concentrate (Davenport et al., 2002). Pyrometallurgical processes are the predominant route to treat copper sulfide minerals. These processes, however, have some problems including the emission of SO_2 and inability to treat complex concentrates and low grade ores. These problems will be more highlighted in the future, because the resources of high grade copper ores in the world are becoming more and more scarce making the processing of more complex ores necessary. During the past decades, attention has been given to hydrometallurgy as an alternative to treat sulfide ores.

Hydrometallurgical processes such as pressure oxidation, bioleaching and chloride leaching have been considered as alternatives for extraction of copper from chalcopyrite flotation concentrates.

Galvanically-assisted atmospheric leaching of copper concentrates is one of the promising technologies for leaching of chalcopyrite concentrate. This technology is based on the anodic dissolution of chalcopyrite (oxidation of sulfur) and cathodic reduction of Fe^{3+} to Fe^{2+} in ferric sulfate media. To consume the produced electrons during chalcopyrite dissolution, pyrite can act as an effective provider (catalysts) for ferric reduction. The key of this process was reported to be the presence of pyrite at a level of typically two to four times the mass of chalcopyrite (Dixon et al., 2008). The main reaction for chalcopyrite dissolution in ferric/ferrous sulphate medium is the oxidation of chalcopyrite (S^{2-}) by ferric ions. Addition of a large amount of pyrite and production of Fe^{2+} can be considered as important limitations of this process, even, when pyrite could be recycled and reused.

During the past 30 years operations using microorganisms for metal recovery have progressed from dump and heap bioleaching of low grade ores to controlled oxidation and bioleaching in designed bioheaps for secondary copper sulfide ores and refractory gold ores and in stirred

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bioreactors for recovery of cobalt and also refractory gold (Brierley and Brierley, 2001; Olson et al., 2003; Rohwerder et al., 2003; Ranjbar et al., 2007; Walting, 2015; Mahmoud et al., 2017). Bioleaching of chalcopyritic concentrate is on the verge of commercialization. In this regard, many researchers in universities and industry have focused their efforts on finding a way to extract copper from chalcopyrite by bioleaching (d'Hugues et al., 2008; Petersen and Dixon, 2006; Cordoba et al., 2008; Hiroyoshi et al., 2008; Kauber, 2008; Gericke et al., 2008; Ahmadi et al., 2010; 2011; Chemielewski and Kaleta, 2011; Majuste et al., 2012; Vakylabad et al., 2012; Lotfalian et al., 2015, Khoshkhoo et al., 2017, Esmailbeigie et al., 2017). From the literature results, it can be pointed out that the main hindrance to achieve an efficient bioleaching process for chalcopyrite is its low dissolution rate even at highly acidic conditions. It has been also shown that the one important reason for this behaviour is the surface passivity of chalcopyrite by a controversial inhibiting layer at high solution potentials which acts as a diffusion barrier for leaching agents and reaction products. The most often suggested oxidation products for the formation of passive layers are elemental sulfur and jarosite. To overcome the problem, the formation of the passive layer must be prevented by optimizing electrochemical, physicochemical, biological and operational parameters. The most promising strategy for improved chalcopyrite bioleaching has been reported to be the possible control of pulp ORP (oxidation reduction potential) during the bioleaching process. It was also reported that the addition of carbon and pyrite can increase the galvanic interaction between the carbon or pyrite as cathode and chalcopyrite as anode. As a result of this interaction the formation the passive layer could be reduced, which lead to an increased chalcopyrite dissolution (Ahmadi et al., 2012; 2013).

During microbial assisted electrochemical leaching of copper sulfide minerals (chalcopyrite), iron-oxidizing microorganisms catalytically generate the necessary leaching agents (Fe^{3+}) and sulfur oxidizing microorganisms can oxidize elemental sulfur during an acid production process. Furthermore, as a results of galvanic interactions between pyrite and chalcopyrite, pyrite particles (with the higher rest potential) can act as a cathode and are protected whereas dissolution of chalcopyrite particles (with the lower rest potential (as anode) is increased. It is believed that the generated electrons during biooxidation of Fe^{2+} to Fe^{3+} and S^{2-} to elemental sulfur would be transferred to the surface of pyrite and could reduce Fe^{3+} to Fe^{2+} and would also react electrochemically with oxygen in acidic media (H^+) to water. Therefore, pyrite is essential during the successful operation of the so called microbially assisted electrochemical (galvanic) leaching of chalcopyrite concentrates. This study was aimed to outline the possible application potential of this novel technology in the copper mining industry.

2. Materials and methods

2.1. Materials

Representative chalcopyrite-rich flotation concentrate samples for this study were obtained from the main Iranian copper processing complexes, Sarcheshmeh, Kerman. The samples were dried and blended proportionally by weight to produce the final sample. The final sample was milled to the target D_{80} of around 45, 16 and 9 (*Fig. 1*). The results of process mineralogy analysis of feed (flotation concentrate) showed chalcopyrite (59.7 wt%) and pyrite (23.4 wt%) as the main phases and covellite (4.7 wt%), chalcocite (0.9 wt%), bornite (1.4 wt%), copper oxides (0.3 wt%) sphalerite (1.4 wt%), molybdenite (0.2%) and other non-copper minerals mainly silicates as minor phases. Chemical analysis by X-ray fluorescence (XRF) showed that the concentrate included 26.2% Cu, 29.7% Fe and 36.1% S. Pure crystalline pyrite hand selected samples from Sarcheshmeh mine were used in experiments with pyrite addition. These samples were washed using a solution (10% H_2SO_4) for removal of acid soluble gangues and milled to the target D_{80} of 20 μm (*Fig.1*). Dolomite (7.8 wt%) was identified as the minor phase of the

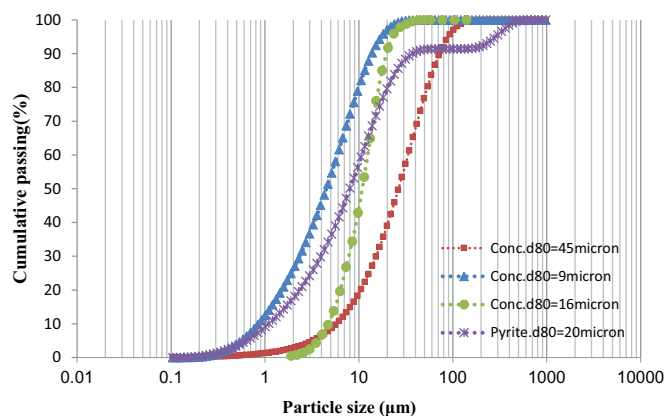


Fig. 1. Particle size distribution of the copper concentrates and pyrite

pyrite.

2.2. Cultures and nutrient media

A mixed culture of moderately thermophilic iron- and sulfur oxidizing bacteria mainly containing *Acidithiobacillus caldus thermosulfidooxidans* obtained from Sarcheshmeh Biohydrometallurgy Laboratory and a mixed culture of *Thermoplasma acidophilum* and *Acidianus brierleyi* received from Federal Institute for Geosciences and Natural Resources (BGR, Hannover, Germany) were used as inoculum. The cultures were initially grown in shake flasks using a temperature controlled incubator at 50 °C, pH = 1.2 and 180 rpm in modified 9 K medium which consisted of 3.2 g/L $(\text{NH}_4)_2\text{SO}_4$, 0.76 g/L K_2HPO_4 , 0.3 g/L MgSO_4 , 0.08 g/L KCl, 0.02 g/L $\text{Ca}(\text{NO}_3)_2$. As energy sources, 3% elemental sulfur and 20 g/L ferrous sulfate were added to the medium. Beginning with a solid content of 1%, the cultures were stepwise adapted to copper concentrate at pulp densities up to 15%. The adaptation process was repeated until a bacterial density of around 10^9 cells/ml was achieved within 3 days. This process was monitored using Eh and pH data as a function of adaptation time as well as cell densities (cells/ml).

2.3. Analytical techniques

The mineral and chemical composition of the chalcopyrite concentrate, hand selected pyrite samples and leach residues were determined by phase analysis and chemical element analysis techniques, respectively. The mineral species were analyzed using Leica phase contact microscope (British Standard 3406-part4) and a D/max-3B X-ray powder diffractometer through Cu K α radiation (XRD). Scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM/EDS) and X-ray fluorescence (XRF) was used to determine the chemical composition of solids. The copper and iron concentrations in pregnant leach solutions (PLS) were measured by atomic absorption spectroscopy in the liquid fraction from centrifuged pulp samples.

2.4. Apparatus and procedures

The leaching experiments were performed in a continuously operated system which consisted of three stirred tank reactors (CSTR). The feed circuit consisted of an agitated slurry feed/pump tank to which inoculum could be added together with milled samples to produce the target solid density of 10 w/w %. The slurry feed was then pumped from tank to tank using a peristaltic pump. To prevent the undesirable change of process parameters, all tanks were equipped with external heating and monitoring instrumentation (pH, T, ORP and aeration system). Three consecutive reactors had the following volumes and residence times: Reactor 1: 5 L, 42 h; reactor 2: 5 L, 42 h; and reactor 3:

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