



A conceptual process for copper extraction from chalcopyrite in alkaline glycinate solutions



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ABSTRACT

A conceptual flowsheet is proposed and the main processing steps are evaluated for the alkaline processing of chalcopyrite where glycine is the complexing agent. Glycine is utilised in an oxidising, alkaline environment to leach chalcopyrite at atmospheric pressure and mildly elevated temperatures. Process steps to recover copper and glycine from alkaline aqueous solutions were also investigated. The leaching of chalcopyrite flotation concentrate in glycine solutions was conducted at different leach conditions in a 1.25 L leach reactor with an agitated slurry and controlled dissolved oxygen (DO) concentration. In the presence of air, oxygen or hydrogen peroxide or a mixture thereof, glycine can dissolve copper from chalcopyrite at either ambient or elevated (40–60 °C) temperatures and atmospheric pressure. Increasing temperature, pH, glycine concentration and DO concentration all increase the rate and extent of copper extraction. The extraction of copper from “as-received” chalcopyrite flotation concentrate, at a particle size of 100% –45 µm, in solutions containing 0.4 M glycine at 60 °C with 25 ppm DO, was 40.1% after 24 h. If the chalcopyrite concentrate underwent an ultrafine grind to 100% –10 µm with prior alkaline atmospheric pre-oxidation, 92% of the copper is leached within 17 h at 60 °C, atmospheric pressure and 9% solids during a batch leach. Pyrite associated with the chalcopyrite remained unreacted during leaching of chalcopyrite and iron concentration in the final pregnant solution was found to be less than 20 mg/L. Copper recovery by sulfide precipitation from the leach solution as pure covellite was up to 99.1% at a Cu:S²⁻ molar ratio of 1:1. Solvent extraction (SX) experiments with LIX 84-I demonstrated that copper can be extracted into the organic phase up to 99.4% in a single stage at an equilibrium pH range 8.8–10.0. It is shown that copper can be stripped from the organic phase in a single stage acidic strip using conventional acidic return electrolyte containing 180 g/L sulfuric acid. During copper recovery by precipitation as sulfide, or by solvent extraction, the glycine is made available for recycling and reuse as a barren leach solution, after treatment with lime.

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1. Introduction

1.1. Background to chalcopyrite leaching

The objective of this research was to evaluate a proposed conceptual flowsheet for the leaching of chalcopyrite using aqueous glycine in an alkaline environment with a suitable oxidant, and to identify potentially feasible routes for copper recovery from such a pregnant leach solution with concomitant reagent regeneration. The approach is based on earlier research by the authors (Oraby and Eksteen, 2014) that indicated that chalcopyrite from

copper-gold gravity concentrates is solubilised in such a system. The scope of the research was to experimentally validate the key metal recovery steps of an integrated process, rather than a fundamental study of each process step, in order to serve as a reference and provide context for further research. Such an approach to chalcopyrite leaching is warranted due to the challenges associated with the treatment of chalcopyrite ores via conventional leaching approaches, as will be discussed below.

The persisting trend of decreasing grades of copper and copper-gold ores, the occurrence of finely disseminated chalcopyrite in gold-bearing pyrite and the presence of deleterious contaminants, limits the extent to which conventional milling and flotation processes can be used economically to produce clean flotation concentrates that are acceptable for smelting. A significant body of research has accumulated over the past two decades on the

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extraction of copper from low grade copper or copper-gold deposits (Carranza et al., 2004; Dixon et al., 2007; Maley et al., 2009; Turan and Altundoğan, 2013a,b). The profitable extraction of copper from low-grade ores requires low-cost processing methods such as in situ or heap leaching (Watling, 2006). Copper (often with precious metals) is predominantly found as chalcopyrite in copper porphyries, iron-oxide-copper-gold (IOCG) and volcanic massive sulfide (VMS) deposits. This study will focus on chalcopyrite as it is the most abundant copper mineral and because of its known refractory nature to conventional acid leaching. The leaching of gold (and non-dissolution of pyrite) in oxidising, alkaline, glycine solutions has already been demonstrated by the authors (Eksteen and Oraby, 2015) who also observed that the presence of copper in solution enhances the gold leaching kinetics. In addition, significant increases in gold dissolution is observed when the gold is leached with glycine, copper and starvation amounts of cyanide.

About 70% of the world's copper resources are present as chalcopyrite (Harmer et al., 2006). Many hydrometallurgical processes have been studied to extract copper from chalcopyrite. None of these processes has reached commercial-scale operation due to a diverse range of challenges such as (Wang, 2005): (1) the formation of a passivation layer on the chalcopyrite surfaces; (2) surplus production of sulfuric acid or elemental sulfur; (3) problems with purification of leaching solutions, (4) issues with recovery of precious metals from the leached residues; and (5) the need for stabilization of the final leaching residue for disposal, or (6) the high capital costs associated with pressure oxidative leach processes. The direct leaching of chalcopyrite flotation concentrates and the subsequent solvent extraction-electrowinning processes of copper cannot economically compete with the smelting of the same concentrates for concentrates that meet smelter quality specifications and where existing smelting capacity is available. Electrowinning is energy intensive whereas smelting utilises undiluted material and use the inherent fuel value of the sulfides for the smelting. It is therefore very hard to economically justify chalcopyrite concentrate leaching for concentrates that satisfy the smelter specifications (i.e. "clean" concentrates), particularly in an environment of existing available smelting capacity. However, to obtain high grade "clean" concentrates, the mill-and-float concentrator at the mine often have to reduce flotation mass pulls and suppress (gold bearing) pyrites, implying that a cyanide based tailings leach is often required to recover the gold with concomitant production of significant weak acid dissociable cyanides.

Sulfide concentrates can be treated hydrometallurgically, but leaching of chalcopyrite is difficult and slow and requires strongly oxidising, high temperature or high pressure conditions (Lu et al., 2000; Hiroyoshi et al., 2001; McDonald and Muir, 2007; Yoo et al., 2010), with concomitant impacts on capital and operating costs.

In the field of hydrometallurgy, there are a many publications related to the recovery of copper from chalcopyrite in which different lixiviants such as chloride (Hirato et al., 1986; Liddicoat and Dreisinger, 2007; Al-Harashsheh et al., 2008; Yoo et al., 2010; Miki and Nicol, 2011), sulfate (Munoz et al., 1979; Hirato et al., 1987; Córdoba et al., 2008; Nazari and Asselin, 2009), ammonia (Beckstead and Miller, 1977a and Beckstead and Miller, 1977b; Reilly and Scott, 1977; Turan and Altundoğan, 2013a,b; Nabizadeh and Aghazadeh, 2015) and nitric acid (Habashi, 1999) are used. The use of ammonia is problematic for numerous reasons such as its limitation in recovery and reuse, the limited E_h -pH stability fields of its metal complexes, its volatility (especially at elevated temperature), and numerous health and environmental concerns. As conventional alkaline leach options have been limited in technical and economic

appeal, acid leaching (or acidic heap bioleaching) is the predominant hydrometallurgical route to deal with copper (and chalcopyrite) ores. While sulfuric acid is a relatively low cost commodity, acid leaching of copper minerals creates a number of challenges, and even more so when the ore or concentrate to be leached contains gold which may require alkaline cyanidation of the leach residues.

A few salient aspects of acid leaching are listed here to contrast it to leaching in the alkaline systems: (1) Gangue mineralisation may result in high acid consumption. (2) Acid leaching leads to the formation of elemental sulfur as a by-product which can significantly passivate copper extraction. (3) Depending on leach temperature and acid concentration, it can lead to significant iron co-dissolution and jarosite precipitation with concomitant expensive solid-liquid separation of the iron-rich residues. Cost effective iron removal remains one of the major challenges in acid-based non-ferrous hydrometallurgy. (4) If silver is present in the ore, the silver may be locked into an argento-jarosite crystal lattice. (5) Acid leaching interacts with a number of altered silicates to produce silica gels that causes severe operational problems, particularly in solvent extraction circuits. (6) In addition, many of these silicates can release fluoride or other halide ions in strong acidic environments. Fluorides are also problematic for smelters when these gangue minerals appear in flotation concentrates. (7) Acids mobilise magnesium, calcium, iron, manganese and aluminium ions, which accumulate and have to be managed, as they will influence solvent extraction, and can result in scaling of process equipment and unwanted precipitation throughout the process circuit. This may often lead to challenging water balance issues. (8) Should the copper deposit also contain gold, a significant neutralisation cost is incurred by switching from acidic leaching of copper to the alkaline cyanide leaching of gold. (9) Over and above the raw material costs, particularly if the acid has to be transported over large distances, significant neutralisation costs can be incurred in some instances. The main disadvantages of the alkaline glycine leach system are: it is more expensive than sulfuric acid, ultrafine grinding may be required for high copper extraction and it is oxygen intensive as sulfur is fully oxidised to sulfate (rather than elemental sulfur). The precipitation of gypsum and iron hydroxide may form a surface coating on the copper surface if lime is used as a pH modifier.

Even where smelters with acid production facilities may be at hand to smelt sulfide flotation concentrates, acid production is limited by the overall regional market to absorb the excess acid. This is particularly problematic for inland smelters where copper production can be limited by the ability of the regional market to absorb excess sulfuric acid and large scale storage is a major environmental and safety risk.

Given these constraints related to acid leaching and the operability, health, safety and environmental constraints of other alkaline routes (cyanide and ammonia leaching), other more benign alternatives were considered as candidates for leaching chalcopyrite in the alkaline pH region.

A conceptual process is proposed below which involves leaching copper from chalcopyrite in an alkaline glycine solution at room or elevated temperature (40–60 °C) using air, or oxygen, or hydrogen peroxide, or a mixture of these as an oxidant in the leach system. In recently published research work, the authors have developed a process using an alkaline glycine system to leach copper from a range of oxide (Tanda et al., 2017a) and sulfide minerals, as well as native copper (Oraby and Eksteen, 2014). Additionally, the authors have also shown that this leaching system is applicable to gold and silver (Eksteen and Oraby, 2015; Oraby and Eksteen, 2015a). It has also been demonstrated by Oraby and Eksteen (2015b) that

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