



## Leaching of chalcopyrite ore agglomerated with high chloride concentration and high curing periods



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### ABSTRACT

It is well known that the leaching of chalcopyrite has very slow kinetics. The generally higher reactivity of chalcopyrite in chloride as opposed to sulfate solutions has resulted in many studies of chloride systems for the hydrometallurgical treatment of chalcopyrite. Despite much progress on the chloride leaching process, agglomeration and curing has received less attention over the past years. This paper reports a study on the effect of addition of chloride ion in the agglomeration and the effect of curing time on the overall leaching process of chalcopyrite ore. The work involved leaching tests carried out in columns containing 3.0 kg of agglomerated ore irrigated with raffinate solution under ambient conditions. Experiments with agglomerates produced at different concentration of chloride (20, 50 and 70 kg/t) leach at approximately the same rate. It has been demonstrated that chloride ions presence is necessary to improve the leaching kinetics, but high concentrations of chloride are not essential under ambient conditions. The rate of dissolution of chalcopyrite ore agglomerated with high concentrations of acid and chloride ions increases as the curing period increases from 15 to 80 days observing extraction of copper up to 60% in 20 days of leaching. In all experiments, the shape of the leach curves indicate that the copper that has been solubilized during the curing period is rapidly leached followed by continuous slow leaching thereafter. This study supports the addition of chloride at the agglomeration stage as a viable route for the treatment of low-grade copper sulfide ore.

### 1. Introduction

Currently the Chilean mining industry has to face several problems; among them are the depletion of oxide copper minerals, the shortage of water resources, a projected increase in the generation of waste and high energy costs (Cochilco, 2016 and Ghorbani and Kuan, 2017). It is projected that copper concentrate production in Chile will almost double from 2014 to 2026, being 88% of the national mining production, which means an increase from 3.9 to 5.4 million tons of concentrate (Cochilco, 2016; Cifuentes and Cantallopis, 2016). For this reason, there is increased concern for air pollution and sulfur dioxide emissions from the conventional smelting processes. Consequently, environmental regulations to protect surface and groundwater pollution will be much stricter, which may restrict the treatment of minerals by this route (Jimena, 2006). On the other hand, hydrometallurgy offers an ecologically more acceptable route to process not only oxides but also sulfide ore, especially for low-grade ores (Pradham et al., 2008). Therefore, the development of mining operations in Chile depends on the development of new hydrometallurgical alternatives (Gok

and Anderson, 2013).

Chalcopyrite represents approximately 70% of the world copper mineral reserves (Habashi, 1978; Veloso et al., 2016). Yet it is one of the most refractory to hydrometallurgical processes. For this reason, since the 19th century, 80% of world copper production has been generated by the conventional pyrometallurgical routes, with the remainder relating mostly to heap leaching of copper oxide ores.

One of the most important unresolved problems in the development of a heap leach process for copper sulfide ores is the inhibited leaching of chalcopyrite at ambient temperature. Numerous studies have reported the formation of a layer on the surface of chalcopyrite during the dissolution process; however, the nature of this “passivating layer” has not yet been established with certainty. This phenomenon would be inhibiting the contact of mineral with oxidizing agents, reducing the dissolution rate (Debernardi, 2013; Kawashima, 2013; Viramontes et al., 2010 and Córdoba et al., 2008). In addition, different authors agree that dissolution of sulfide minerals, and in particular the passivation of chalcopyrite, are potential-dependent reactions, and many studies have been devoted to the elucidation of the potentials at which

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the dissolution is possible and at which passivation occurs (Hiroyoshi et al., 2001; Nicol and Lazaro, 2002; Nicol et al., 2010; Nicol and Basson, 2017). These studies established the relevant leaching potential region as being 0.45 to 0.75 V vs SHE. On the other hand, Velásquez-Yévenes et al. (2010a) in leaching experiments conducted at controlled potential and low chloride concentrations, established the enhanced leaching potential region as being 0.55 to 0.62 V vs SHE, but higher concentration of chloride could extend this region to higher values (Velásquez-Yévenes et al., 2010b).

The leaching of chalcopyrite in chloride media presents many advantages in hydrometallurgical processes with respect to sulfate systems. Chloride leaching can be carried out at ambient temperatures, whereas sulfate processes generally require elevated temperatures. In addition, in chloride systems the copper(I) ion is stable (Watling, 2013), and this allows the use of copper(II) ions as an oxidant in addition to iron(III) (Velásquez-Yévenes et al., 2010a). In some instances, greater leach kinetics in a chloride system could be caused by the enhanced proton activity (Senanayake, 2007), in that the addition of chloride solvates free water, causing an increase in the proton activity (Muir, 2002). There is extensive research in leaching with ferric chloride and cupric chloride to treat copper sulfide concentrates (Peters, 1977; O'Malley & Liddell, 1987; Velásquez-Yévenes et al., 2010a, 2010b, Nicol et al., 2010). Chalcopyrite leaching experiments in ferric-chloride media conducted by Dutrizac (1990), found that the surface of the passivating layer formed on the ore was composed of 95% elemental sulfur, under the conditions of the experiments. Lu et al. (2000a, and 2000b) postulated that chloride ions modify the morphology of the deposited sulfur, which is more crystalline and porous, thus permitting the dissolution reaction to proceed. Carneiro and Leão (2007) presented similar findings.

Cordoba et al. (2008 and 2009), in their research found that the nature of the passivation layer formed on the chalcopyrite surface is related to the pH. Authors have shown that the highest rates of leaching were achieved at pH between 1.5 and 2.0, and the passivating layer formed was composed of jarosite from the hydrolysis of iron. In contrast, Velásquez-Yévenes et al. (2010b) reported that the chalcopyrite dissolution rate is independent of pH in the range 0.5 to 2.0, since at these pH values the iron (III) can be kept in solution and its precipitation can be prevented.

Although there is a considerable body of published information on the dissolution of chalcopyrite in chloride solutions, the emphasis has been placed on the leaching process and not on the pretreatment of the crushed ore (agglomeration and curing time) and even less on the possible addition of chloride in the agglomeration stage. There is agreement that in a heap leaching process crushed ore agglomeration and curing time, as pretreatment stages, can be crucial for the success of the overall leaching process (Kodali et al., 2011; Dhawan et al., 2013). However, it seems that agglomeration and curing practice is based largely on experience and lacks a more rigorous scientific understanding (Lu et al., 2017).

If crushed ore that has not been agglomerated is stacked for heap leaching, migration of fine particles with the leaching solution is likely to occur during irrigation. This is likely to reduce the permeability of the heap through blockage and obstruction of the flow channels and pores, resulting in a low extent of dissolution of the ore (Bouffard, 2005; Dhawan et al., 2013; Kodali et al., 2011 and Readett and Fox, 2011). Frequently, the addition of raffinate-based solution alone or in conjunction with other additives, such as Portland cement, bentonite and geopolymers, is used as a media to support agglomeration (Lewandowski and Kawatra, 2009). The role of the agglomeration media is to bind the ore particles together by liquid bridges to increase the permeability of the heap. Depending on the mineralogy of the ore and environmental conditions, the wet agglomerates undergo curing prior to stacking on a heap. Curing times with sulfuric acid have been reported from 15 to 300 h (Dhawan et al., 2013). During curing, the acidic raffinate reacts chemically with soluble particles, generating dry

agglomerate that is enhanced by the recrystallization or solidification of the solubilized species and consequently improves the leaching rate of the ore (Purkiss and Anthony, 2004; Quaiocoe et al., 2015). In addition, curing inhibits the dissolution of some silicate impurities and leads to a homogeneous distribution of the acid in the ore bed along with higher porosity in the bed (Cruz et al., 1980; Jansen and Taylor, 2003).

Cerda et al. (2017) studied the effect of pretreatment on the leaching of a copper sulfide ore in acid-chloride media, indicating that chloride enhanced the dissolution of ore, with maximum extraction of 49% in 78 d of leaching. Velásquez-Yévenes and Quezada-Reyes (2018) found that curing time and an intermittent irrigation regime enhance the dissolution of chalcopyrite ore leached in column. No additional information has been found on the effect of chloride ions on the pretreatment of the chalcopyrite ore and no study dedicated to investigating the effect of different curing periods in the leaching of copper ores was found. Hence, the overall aim of this study is to investigate the effect of the addition of high acid-chloride concentration solution in the agglomerate stage, well as the effect of different curing times on the subsequent leaching of chalcopyrite ore in chloride media under ambient conditions.

## 2. Experimental

### 2.1. Materials

The copper ore sample used for these experiments was provided by a heap leaching operation, located in Antofagasta, Chile. The chemical analysis of the ore at different size fractions is shown in Table 1.

Mineralogical analysis of the ore sample was performed using Quantitative Evaluation of Minerals by Scanning Electron Microscopy (QEMSCAN). The copper-bearing sulfide minerals in the ore consist mainly of chalcopyrite ( $\text{CuFeS}_2$ ), followed by minor chalcocite ( $\text{Cu}_2\text{S}$ ), covellite ( $\text{CuS}$ ), bornite ( $\text{Cu}_5\text{FeS}_4$ ) and traces of other copper minerals (Fig. 1). The complete mineralogical composition of the ore is shown in Table 2.

### 2.2. Agglomeration procedure

Agglomeration tests were conducted in a batch laboratory-scale polypropylene drum of 16 cm diameter with a volume of 5 L through a system of rotating rollers operated at 75 rpm rotational speed. The selection of drum speed was based on preliminary agglomeration tests at 50, 60, 75 and 80 rpm, which showed 75 rpm to be most efficient, taking into consideration the batch time of 5 min required to form the desired agglomerates. For all agglomeration tests, 3 kg of well-mixed crushed ore with a particle top-size of < 6.5 mm was used. Fig. 2 shows the particle size distribution of the sample. Different amounts of chloride (as solid NaCl), acid, and the required amount of water to obtain 10% of humidity were added to the agglomeration. The agglomerates produced were placed in columns and left to rest for specific periods, depending on the experimental condition.

### 2.3. Column leaching tests

Leaching of all agglomerates was carried out in acrylic columns, 87 mm in diameter and 450 mm in height. A perforated acrylic disk was

**Table 1**  
Chemical composition of the ore by ICP-MS.

Size fraction ( $\mu\text{m}$ )	Weight (%)	Cu (%)	Fe (%)	S (%)
–75	38.2	0.3	3.9	0.5
–75 + 38	13.9	0.3	4.2	0.6
–38 + 25	40.9	0.3	4.0	0.4
–25 + 5	7.0	0.2	4.0	0.2
Total (calculated)	100.0	0.3	4.0	0.5

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