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# The effect of seawater based media on copper dissolution from low-grade copper ore

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# ABSTRACT

Due to the scarcity of water in the north of Chile, there is interest in small-scale mining using seawater to leach the minerals. This situation has led to this research aimed to determine the effect of different process variables on the extraction of copper based on the ore type of these small-scale mining activities. The extraction of copper from finely ground (<150  $\mu$ m) low-grade mixed ore (0.36% Cu) was studied in different acidic media (H<sub>2</sub>SO<sub>4</sub> and HCl). The effects of water quality (tap water, seawater and synthetic process water) and lixiviants on copper leaching were investigated at three temperatures (25 °C, 35 °C and 45 °C). Synthetic process water was prepared by adding Na<sup>+</sup>, Cl<sup>-</sup>, Cu<sup>2+</sup> and SO<sub>4</sub><sup>2-</sup> to seawater. Copper extractions between 70% and 80% were achieved in 48 h using seawater, similar to the extractions obtained using tap water. Rapid copper dissolution occurred when synthetic process water was used, from 90% to 97%. This marked increase was related to the addition of Cu<sup>2+</sup>, which promoted the formation of CuCl<sup>+</sup>. Seawater was comparable to freshwater in terms of leaching kinetics and yield potential by raising the chloride concentration and increasing the formation of copper chloride ions. The findings of this study also expanded our understanding of the consequences of substituting seawater for freshwater at industrial leaching operations.

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# 1. Introduction

In recent years, several processes have been proposed for the leaching of sulfide ores and copper concentrates in chloride media: the CLEAR process (Atwood and Curtis, 1974, 1975); the Cuprochlor process (Aroca et al., 2012); the Intec process (Moyes and Houllis, 2002); the CESL process (Barr et al., 2000); the CUPREX process (Dalton et al., 1991) and the HydroCopper process (Hyvärinen and Hämäläinen, 2005). Comprehensive reviews of these processes in terms of their conditions, commercial development and industrial applications have been published, including the work of Dreisinger (2006) or McDonald and Muir (2007a,b), to which the reader is referred.

Chloride systems present a number of advantages and disadvantages (Lu and Dreisinger, 2013a,b; Dutrizac, 1992) compared with sulfate systems (Table 1).

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The utilization of chloride solutions in chalcopyrite leaching is advantageous due to the aggressive nature of the leaching and to the stability of cuprous ions due to the formation of chloro-complex ions. In general, the leaching of chalcopyrite is more effective in chloride solutions with cupric ions as the oxidant than in sulfate solutions with ferric ions as the oxidant. This is possibly due to kinetic rather than thermodynamic considerations; for example, the higher rates of electron transfer in chloride solutions than in sulfate solutions, as the chalcopyrite surface passivates more readily in the presence of sulfate ions (Hackl et al., 1995). The formation of passivating reaction product layers has been suggested several times in sulfate solutions (Hackl et al., 1995; Parker, et al., 2003). Munoz et al. (1979) concluded that the rate-limiting step in the leaching of chalcopyrite with ferric sulfate was the transport process through the sulfur product layer. Similar behavior has also been reported in ferric chloride leaching (Munoz et al., 1979).

Lu et al. (2000) focused their electrochemical studies on the leaching of chalcopyrite in mixed sulfate–chloride media and found that the presence of chloride enhanced chalcopyrite





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oxidation. Kinnunen and Puhakka (2004) reported that the presence of 0.25 g/L of Cl<sup>-</sup> in leaching media enhanced the copper yield at temperatures between 67 and 90 °C but decreased the copper vield at 50 °C. Nicol and co-workers (Miki and Nicol, 2011; Miki et al., 2011; Nicol et al., 2010; Velásquez-Yévenes et al., 2010a,b) conducted a wide ranging study of the dissolution of chalcopyrite in chloride solutions containing cupric ions and dissolved oxygen. Initially, it was shown that the rate of chalcopyrite dissolution was enhanced when leaching was conducted in an oxidation reduction potential (ORP) range of 550-620 versus a SHE at 35 °C in a solution containing 0.2 M HCl, 0.008 M Cu<sup>2+</sup> (simulating raffinate) and 5–15 mg  $L^{-1}$  dissolved O<sub>2</sub> (Velásquez-Yévenes et al., 2010a). Leaching at ORP < 540 mV caused reduced rates of chalcopyrite dissolution and covellite or chalcocite formed on some chalcopyrite surfaces. Operations at locations with higher temperatures or the development of methods to economically heat the solutions to above 25 °C may be a prerequisite for the successful implementation of chloride leaching of ores containing primary copper sulfide minerals (Basson et al., 2010).

The leaching kinetics of copper sulfides in ferric and cupric chloride solutions depend on the cupric ion to cuprous ion ratio as well as the ferric ion to ferrous ion ratio. Parker et al. (1981) attributed faster chalcopyrite oxidation rates in mixtures of 0.5 M CuCl<sub>2</sub> and 0.5 M FeCl<sub>3</sub> to high corrosion potentials generated at the mineral surface because the CuCl<sub>2</sub> reduction at the chalcopyrite surface is faster than the FeCl<sub>3</sub> reduction; they suggested that the role of FeCl<sub>3</sub> was to minimize the concentration of chloro-complex CuCl<sub>2</sub>. Further details of the chalcopyrite leaching in chloride have been discussed in the review by Watling (2014).

Seawater presents an alternative to freshwater and has been used at some mine sites. For example, BatuHijau (Indonesia), TexadaMill, Raglan (Canada), BHP Nickel (Australia), Lipesed, Michilla, Esperanza, Algorta Norte, Mantos de la Luna and Las Luces (Chile) have used raw seawater in their processes (González et al., 2012; Philippe, 2012; Moreno et al., 2011; Aral et al., 2010; Quinn et al., 2007). In the case of Chilean copper mining, mineral deposits are found mainly in the north of the country where the weather is arid with sparse or no rainfall. In general, the Antofagasta region is responsible for 36% of the production of concentrates and for 72% of the production of copper cathodes at the national level; therefore, the region has high water consumption. Eighteen percent of the water used in mining processes in this region comes from seawater (COCHILCO, 2013). Seawater could be beneficial to the hydrometallurgical processing of copper sulfides, particularly of chalcopyrite due to the presence of chloride ions. Moreover, leaching with seawater has been studied previously (e.g. Torres et al., 2013; Hernández et al., 2012; Taboada et al., 2012; Glasby and Schulz, 1999; Beverskog and Puigdomenech, 1998; Plyasunova et al., 1997; Nila and Gonzalez, 1996; Winand, 1991; Berger and Winand, 1984; Fritz, 1980; Bianchi and Longhi, 1973). Due to the shortage of water in the north of Chile, there is interest in smallscale mining using seawater to leach the minerals.

#### Table 1

lvantages/disadvantages of chloride systems.	
Advantages	Disadvantages
<ul> <li>The increased solubilities of iron and other metals</li> <li>The enhanced redox properties because cupric and cuprous ions were stabilized as chloride complexes and the Cu(1)/Cu(II) redox couple could contribute to sulfide oxidation reactions</li> <li>The faster leaching kinetics of chalcopyrite compared with sulfate systems</li> <li>The generation of elemental sulfur rather than sulfate</li> <li>Low pyrite reactivity in chloride systems</li> </ul>	<ul> <li>The corrosive action of chloride, thus necessitating the use of more expensive materials of construction for reactors</li> <li>The need for fine grinding for processes operated at atmospheric pressure</li> <li>The co-leaching of multiple elements that require additional treatments</li> <li>The difficulty of electrowinning high-grade copper from chloride solutions</li> </ul>

The Laboratorio de Investigacion de Procesos (LIP) at the University of Antofagasta (UA) hasconducted a broad range of research studies (Hernández et al., 2012; Taboada et al., 2012; Torres et al., 2013) with regards to seawater applications in the mining industry. Understanding the effect of using seawater on ore beneficiation processes from technical, economical and environmental points of view is the main objective of the water research in LIP at UA.

In the present study, the effect of sea water-based media on copper dissolution from low-grade ore was investigated. Experiments were performed at three temperatures (25, 35 and 45 °C) using H<sub>2</sub>SO<sub>4</sub> and HCl as lixiviants and seawater, tap water and process water as solvents. In addition, this work assesses the potential benefit of cupric ion and copper chloride complexes in the dissolution of copper using different media containing chloride and cupric ions.

# 2. Materials and methods

### 2.1. Ore sample

The low-grade copper ore sample was obtained from the Lomas Bavas mining company located close to Antofagasta-Chile. The chemical composition (Table 2) using inductively coupled plasma atomic emission spectroscopy (ICP-AES) showed 0.36% Cu in the ore sample. Major sulfide minerals inferred from the quantitative evaluation of the minerals by scanning electron microscopy (QEM-SCAN) analysis of the feed sample included pyrite (55.9 wt.% of the sulfide components), chalcocite (15.3 wt.%) and bornite (11.3 wt.%) with lesser amounts of covellite and chalcopyrite. Table 3 shows the mineralogical composition of the feed. Due to the oxidized nature of the ore sample, oxide minerals were not identified by QEMSCAN but the study of the selected samples using optical mineralogy showed the presence of copper oxide as atacamite  $(Cu_2Cl(OH)_3)$ and/or antlerite (Cu<sub>3</sub>(SO<sub>4</sub>)(OH)<sub>4</sub>.

#### 2.2. Seawater

The seawater sample was obtained from San Jorge bay, Antofagasta, Chile. Possible insoluble particulate matter and microorganisms in the seawater sample were removed using two stages of filtering: (i) with quartz sand (50  $\mu$ m) and (ii) by passage through a polyethylene membrane (1-µm pore size). Synthetic leaching solution, which is similar to industrial process water, was prepared by adding Na<sup>+</sup>, Cl<sup>-</sup>, Cu<sup>2+</sup> and SO<sub>4</sub><sup>2-</sup> to seawater. As show in Table 4, different analytical techniques including volumetric, gravimetric and instrumental analyses (atomic absorption spectrometry (AAS) and ICP-AES) were used to determine the chemical compositions of the seawater and synthetic process water.

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