



The mechanisms of improved chalcopyrite leaching due to mechanical activation



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ABSTRACT

Chalcopyrite leaching has attracted much attention in both research work and industrial implementation due to its low cost. However, the slow Cu leaching rate during chalcopyrite dissolution process limits its wide application. This paper investigated the influence of mechanical activation on chalcopyrite leaching. The results showed that co-grinding of chalcopyrite and oxidants (*i.e.* $\text{Fe}_2(\text{SO}_4)_3 \cdot x\text{H}_2\text{O}$) stimulated solid-state reactions to form dissolvable copper sulfate, thereby increasing Cu extraction rate significantly. The reactions gradually proceeded with the increase of grinding time and speed, as well as the amount of oxidant. After activation, more than 98% of chalcopyrite was extracted in pH 1.0 sulfuric acid maintained at 348 K for 1.5 h, significantly greater than that without activation, indicating a promising strategy for high efficient chalcopyrite leaching.

1. Introduction

Chalcopyrite (CuFeS_2) is one of the most abundant and widespread minerals containing copper (Harmer et al., 2006), accounting for almost 70% of the copper resources in the world (Li et al., 2013). Although pyrometallurgical processes are currently producing approximately 80% of copper (Watling, 2013), it can only be economically used for high grade copper minerals. However, with increasing imbalance between copper supply and world demand, high-grade copper ores are gradually decreasing and the processing of low-grade ores should be considered urgently (Watling, 2013). In addition, pyrometallurgical processes of chalcopyrite emit SO_2 gas, resulting in serious air pollution (Juarez et al., 2006).

In recent years, much attention has been paid to hydrometallurgical processing strategy which normally uses sulfuric acid, nitric acid and hydrochloric acid as leaching lixiviants (Aguirre et al., 2016; Hu et al., 2017; Khoshkhou et al., 2017; Nicol et al., 2010; Qian et al., 2014; Ruiz-Sánchez and Lapidus, 2017; Veloso et al., 2016). However, most studies have suggested that the leaching process usually leads to slow copper recovery and low copper extraction due to surface passivation (Córdoba et al., 2009; Carneiro and Leão, 2007; Hackl et al., 1995; Qian et al., 2014; Stott et al., 2000; Viramontes-Gamboa et al., 2010).

Many studies have been conducted to improve chalcopyrite leaching rate and the final copper recovery, *e.g.* Pan et al. (2012) reported that copper extraction was increased from 50% to 89.8% by adding glass beads, although jarosite precipitates and weak passivation were still

observed on chalcopyrite surface. Sandström et al. (2005) increased chalcopyrite leaching efficiency by adjusting Fe^{2+} and maintaining the potential at about 450 mV. Other studies adjusting the leaching systems including pH, redox potential, pulp density, temperature, galvanic interaction, pre-treating chalcopyrite surface, have been intensively carried out (Aguirre et al., 2016; Debernardi and Carlesi, 2013; Falagan et al., 2014; Ghahremaninezhad et al., 2013; Hernández et al., 2015; Hu et al., 2017; Khoshkhou et al., 2017; Khoshkhou et al., 2014; Ruiz-Sánchez and Lapidus, 2017; Tao and Dongwei, 2014; Veloso et al., 2016; Watling, 2013). Although these methods have reduced chalcopyrite passivation to some extent, chalcopyrite leaching rate was still very low (*e.g.* more than days) while the copper recovery was expected to increase.

Different from the above methods, the application of mechanical activation processing was proposed as one suitable option to solve chalcopyrite leaching problems. Mechanical activation involves chemical and physicochemical transformations of substances, specifically mechanical activation of sulfides can increase reaction rates and improve leachability of the target minerals significantly (Bafghi et al., 2010; Baláž et al., 2005; Basturku et al., 2017; Kammel et al., 1987; Maurice and Hawk, 1998; Tan et al., 2017; Xu et al., 2017). Several studies have investigated the effects of mechanical activation on various sulfides (Agnew and Welham, 2005; Bafghi et al., 2013; Baláž and Achimovičová, 2006; Maurice and Hawk, 1998; Mohammadabad et al., 2016), indicating its advantages over the traditional leaching methods. For instance, Li et al. (2017) reported that co-grinding of Mo/V sulfides

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and Na_2CO_3 /oxidants in mechanical activation manner formed molybdate and vanadate, achieving an easier recovery of both Mo and V. Baláz and Achimovičová (2006) found that mechanical activation had a positive influence on alkaline leaching of complex sulfides, e.g. the recovery of Sb and As increased 5 times for jamesonite, 1.4 times for enargite and 4 times for tetrahedrite, respectively. Mohammadabad et al. (2016) reported that the copper extraction was increased significantly after a mechanical activation treatment. Bafghi et al. (2013) demonstrated that the leaching rate of chalcopyrite was greatly enhanced by intensive milling by around five orders of magnitude. Given the efficiency, the mechanical activation strategy at room temperature and atmospheric pressure has been considered as alternatives to other high energy consumption methods such as high pressure autoclave leaching.

Although many studies have been carried out to investigate the mechanical activation effects on various minerals, the activation mechanisms of chalcopyrite have not been fully understood, particularly the effects of co-grinding chalcopyrite with oxidants. In the present study, we investigated the co-grinding and leaching mechanisms of chalcopyrite using the most common oxidant ferric sulfate that normally used in the solution leaching systems, e.g. the solid state reaction, mineral phase transformation and surface morphology, thereby identifying the contributors that can significantly improve chalcopyrite leaching due to mechanical activation.

2. Experimental procedures

2.1. Materials and chemicals

Bulk chalcopyrite sample originally from Sonora, Mexico was purchased from GEO discoveries, Australia. The samples were ground to a particle size of +38–75 μm , followed by wet sieving. Subsequently, sonication was applied to remove the clinging fine particles until clear supernate was observed. The resulting samples were oven dried at 30 °C, stored in a freezer to minimize surface oxidation by air before use. The elemental composition of chalcopyrite was determined by chemical titration method, showing 30.20% Cu, 31.59% S, 29.30% Fe. X-ray diffraction (XRD) analysis of the chalcopyrite powder (Fig. 1a) shows a high purity and well crystallized chalcopyrite with a small amount of fluorapatite as an impurity mineral.

2.2. Milling

Analytical grade iron sulfate hydrate ($\text{Fe}_2(\text{SO}_4)_3 \cdot x\text{H}_2\text{O}$) used as the oxidant was purchased from Sinpharm Chemical Reagent Co. Ltd. Grinding device used in this work was a planetary ball mill (Pulverisette-7, Fritsch, Germany, 740 W) consisting of a pair of pots made of ZrO_2 with an inner volume of 45 mL. The ZrO_2 grinding media

were mono-size spheres having a diameter of 15.9 mm.

A total of 2.0 g of chalcopyrite (+38–75 μm) and oxidant ($\text{CuFeS}_2\text{-Fe}_2(\text{SO}_4)_3 \cdot x\text{H}_2\text{O}$) with various mass ratios, 1:0 (no oxidant added), 1:1, 1:2.5, 1:4 and 1:7 were ground for various periods at different mill speeds. Influencing details of these factors were described in Section 3.

2.3. Leaching

Leaching tests using magnetic stirring apparatuses (DF-101S, constant temperature heating magnetic stirrer, Shanghai Keer Instrument Equipment Co., LTD) were performed in a 50 mL glass beaker with 25 mL solution adjusted to pH 1–7 using diluted analytical sulfuric acid ($0.1 \text{ mol}\cdot\text{L}^{-1}$) and sodium hydroxide ($0.1 \text{ mol}\cdot\text{L}^{-1}$) and 0.125 g chalcopyrite in a water bath, with a stirring rate of 200 rpm. Leaching was conducted from 318 K to 348 K for 0.5 h–2 h to investigate the influence of leaching time and temperature. Subsequently, 5 mL solution was collected, filtered with a disposable syringe and 0.45 μm membrane. The copper concentration was determined using a flame atomic absorption spectroscopy using Agilent AA240FS (Agilent Technologies Inc., Santa Clara, CA, USA).

2.4. Characterization

The mineral phases of unactivated and activated chalcopyrite were determined by X-ray diffraction (XRD) ($\text{Cu K}\alpha_1$, D8 Advance, Bruker, Germany). Field emission scanning electron microscopy (Zeiss Ultra Plus, Germany) was used to determine the surface morphology of the ground samples. The solution pH was measured using pH meter (pH 2100, Eutech Instruments, America).

3. Results and discussion

3.1. Effects of mass ratio and mill speed

Fig. 2 shows the copper extraction of ground samples with a mass ratio of 1:0, 1:1, 1:2.5, 1:4 and 1:7 at different milling speeds for 1 h. Chalcopyrite leaching was carried out in pH 1 H_2SO_4 solution at 348 K for 2 h. It is observed that without oxidant addition, the ground chalcopyrite exhibited a very low extraction (i.e. 8%) after leaching, whereas the ground mixture with a mass ratio of 1:1 remained at a level about 40% at all milling speeds. However, copper extraction increased rapidly with a mass ratio of 1:2.5. In addition, copper extraction increased with increasing milling speed under this condition, e.g. approximately 74% copper extraction was achieved at 600 rpm. When the oxidant to chalcopyrite ratio was continuously increased to 4:1 and 7:1, the copper extraction was increased to 82.22% and 90.32%, respectively although a similar value was found at 450 rpm (around 73%) and

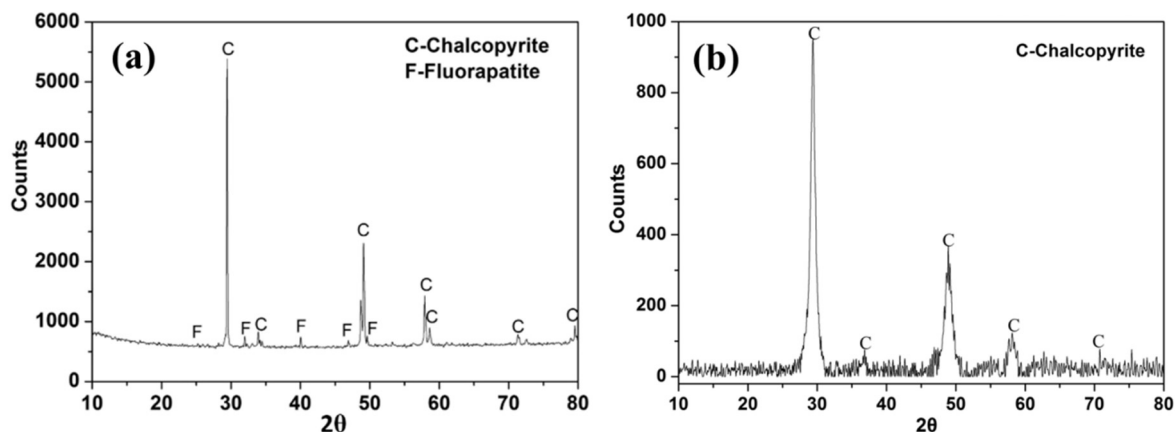


Fig. 1. XRD patterns of chalcopyrite: (a) unactivated and (b) mechanically activated at 600 rpm for 2 h.

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