



MME SEE

CONGRESS 2023

5th Metallurgical & Materials Engineering
Congress of South-East Europe
Trebinje, Bosnia and Herzegovina
7-10th June 2023

CONGRESS PROCEEDINGS

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PROCEEDINGS**

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The Association of Metallurgical Engineers of Serbia

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Institute for Technology of Nuclear and Other Mineral Raw Materials in Belgrade, Serbia;

The Faculty of Technology and Metallurgy at the University of Belgrade, Serbia;

The Faculty of Technology at the University of Banja Luka, Bosnia and Herzegovina;

The Faculty of Metallurgy at the University of Zagreb in Sisak, Croatia;

The Faculty of Natural Sciences and Engineering at the University of Ljubljana, Slovenia;

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A NOVEL APPROACH TO COPPER LEACHING FROM CHALCOPYRITE WITH SULPHURIC ACID SOLUTIONS

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Chalcopyrite is the most abundant copper sulphide mineral. Extensive research in the field of hydrometallurgy was conducted with the aim of developing efficient processes for leaching copper from chalcopyrite concentrates and low-grade ores and/or complex chalcopyrite-containing ores. Chalcopyrite leaching in sulphuric acid solution was most studied with Fe (III) ions as an oxidant, with or without the assistance of microorganisms due to the simplicity and low cost of the process.

In recent years, there has been increased interest in the possible use of different oxidizing reagents (hydrogen peroxide, ozone, chromium (VI) ions, etc.) for chalcopyrite leaching, with research being conducted at the laboratory level, probably due to their higher cost. The addition of organic compounds, such as alcohols (methanol, ethanol, ethylene glycol, isopropanol), in the acidic aqueous solution of hydrogen peroxide and sulfuric acid, had increased chalcopyrite leaching. The aim of this paper is to present a brief overview of the oxidative leaching of chalcopyrite by means of hydrogen peroxide in sulphuric acid solutions and with the presence of alcohols.

Keywords: leaching, chalcopyrite, sulphuric acid, hydrogen peroxide, alcohols

Introduction

The demand for copper has increased significantly in the last two decades. A large part of the world's economic copper resources occurs in the form of chalcopyrite (CuFeS_2), usually with a low content (≤ 0.5 wt.% Cu). As a consequence, the extraction of copper from low-grade sulphide ores is considered economical due to the depletion of high-grade copper ores. Chalcopyrite (CuFeS_2) is a copper iron sulfide mineral consisting of 34.6 wt.% Cu, 30.5 wt.% Fe and 34.9 wt.% S. It is considered the most abundant copper sulfide mineral in most porphyry deposits, which account for about 70 % of the world's Cu resources [1]. Chalcopyrite occurs with other minerals such as pyrite (FeS_2), bornite (Cu_5FeS_4), sphalerite (ZnS), chalcocite (Cu_2S) and covellite (CuS) [2].

Copper extraction from chalcopyrite is mainly carried out by pyrometallurgical (refining, smelting, casting and finally electrolytic refining stages) and hydrometallurgical processes (leaching, solvent extraction and electrowinning). In recent years, extensive research has been carried out in the field of hydrometallurgical processes that can be used for the treatment of low-grade ores and sulphide concentrates without the emission of sulfur dioxide [3].

The largest number of studies on chalcopyrite leaching were performed in sulfuric acid solutions [4-6], hydrochloric acid [3, 7] or nitric acid [8], usually in the presence of various oxidizing agents: (O_2 , Fe^{3+} , Cl_2 , H_2O_2 , oxidants-bacteria, etc.) [9-11]. The main goal of the research is to investigate the kinetics and mechanism of chalcopyrite oxidation, with the aim of developing commercial and cost-effective processes for extracting copper from increasingly low-grade raw materials.

In the last 50 years, the oxidative leaching of chalcopyrite with Fe(III) in sulfate and chloride media has been extensively investigated [11]. Although the leaching of chalcopyrite with iron(III)-sulfate has several advantages (a cheap process with minimal corrosion problems) [12], high recoveries cannot be obtained in this process due to the formation of a layer of reaction products on the surface of the mineral. In addition to the elemental sulfur on the surface of the leached chalcopyrite, other products can also be formed. Mateos et al. [13] indicate that during the leaching of chalcopyrite in an iron(III)-sulfate solution, the passivating layer consists of sulfide (or polysulfide) that is formed by the transformation of chalcopyrite during reaction.

The formation of a passive layer on the surface of the mineral hinders the contact between the mineral and the leaching solution and leads to a reduction in metal extraction.

Given that the leaching rate of chalcopyrite is much lower compared to other sulfide minerals, due to its special crystal and electronic structure [10], finding conditions for more efficient dissolution of chalcopyrite attracts great attention of researchers.

At the laboratory level, research was carried out and strong oxidizing agents were used for chalcopyrite dissolution, such as hydrogen peroxide (redox potential 1.77 V, SHE). The oxidation of chalcopyrite with hydrogen peroxide was mainly investigated in combination with sulfuric acid [9, 14-15]. In order to increase the efficiency of chalcopyrite leaching through the influence on the persistence of hydrogen peroxide in the solution, different organic compounds are applied and their influence on the mechanism and kinetics of mineral oxidation is examined [16-20].

Leaching in the presence of organic acids and alcohols

Recent research studies have shown that the addition of short-chain organic compounds such as methanol, ethanol, ethylene glycol, acetone, isopropanol and/or carboxylic acids (formic and acetic) in acidic solutions ($\text{H}_2\text{SO}_4\text{-H}_2\text{O}_2$) significantly improves the dissolution of chalcopyrite [21].

The dissolution of chalcopyrite was investigated in the presence of hydrogen peroxide and ethylene glycol [22]. The addition of small amounts of ethylene glycol was observed to significantly improve copper extraction. The mechanism of chalcopyrite dissolution was determined based on the examination of various parameters. The effect of temperature on the reaction rate indicates that the surface reaction controls the overall leaching rate. The reaction order calculated in relation to the concentration of hydrogen peroxide is equal to 1 indicating that the concentration of peroxide has a greater influence on the leaching rate. It was found that elemental sulfur is present on the surface of chalcopyrite and that the addition of ethylene glycol stabilizes hydrogen peroxide at higher temperatures, which can later affect the improved extraction of copper [22].

Ruiz-Sánchez and Lapidus [18] also examined the effect of adding ethylene glycol on the leaching of chalcopyrite concentrate in the acidic $\text{H}_2\text{SO}_4\text{-H}_2\text{O}_2$ solution. For this purpose, the chalcopyrite concentrate was leached at a temperature of 20 °C and atmospheric pressure (79 kPa). The test results showed that in the absence of ethylene glycol decomposition of hydrogen peroxide occurs (about 99 % of the peroxide decomposes after 24 h) and that this decomposition is catalyzed by Fe^{2+} , Fe^{3+} and Cu^{2+} ions (Fenton reagents) that are present in the solution as a consequence of the mineral dissolution process itself. The hydroxyl radicals, which are formed as products in Fenton's reactions, can lead to the appearance of peroxide decomposition. After 24 h of leaching, copper and iron extraction ceases (44 % Cu and 37 % Fe extracted). In contrast, in the presence of ethylene glycol, the peroxide consumption was minimal (~16 %) in the same time period and the final extractions for copper and iron were 90 and 71 %, respectively.

The use of organic acids and alcohols in the process of chalcopyrite oxidation in sulfuric acid solutions was the topic of research by the authors Solis-Marcial and Lapidus [16-17]. The electrochemical and chemical dissolution of chalcopyrite in the presence of formic and acetic acids, as well as in the presence of methanol and ethanol was examined [16]. Chemical tests of chalcopyrite concentrate leaching were performed in a solution of $1.0 \text{ mol/dm}^3 \text{ H}_2\text{SO}_4$ (with and without organic acids) in the presence of $0.2 \text{ mol/dm}^3 \text{ CuSO}_4$ and $0.035 \text{ g O}_3/\text{min}$. The experiments were performed at a temperature of 40 °C. The copper extraction values obtained in the presence of formic acid are approximately the same as the copper extraction values obtained in experiments with pure sulfuric acid. This result is probably due to the decomposition of formic acid by oxidants (CuSO_4 and O_3) during the reaction.

On the other hand, it was shown that in the presence of acetic acid, copper extraction reaches 65 % during 5 h of leaching with a tendency to further increase with time.

The results have shown that the kinetics of chalcopyrite oxidation in solutions of $1.0 \text{ mol/dm}^3 \text{ H}_2\text{SO}_4$ with alcohols is faster compared to the kinetics of oxidation in solutions of $1.0 \text{ mol/dm}^3 \text{ H}_2\text{SO}_4$ with organic acids, as well as compared to the kinetics of oxidation in a solution of $1.0 \text{ mol/dm}^3 \text{ H}_2\text{SO}_4$ [16].

The positive effect of the methanol on acidic chalcopyrite leaching was shown by Petrović and Bogdanović [23]. The results obtained on chalcopyrite leaching with hydrogen peroxide in sulfuric acid solution in the presence and absence of methanol are presented in Figure 1.

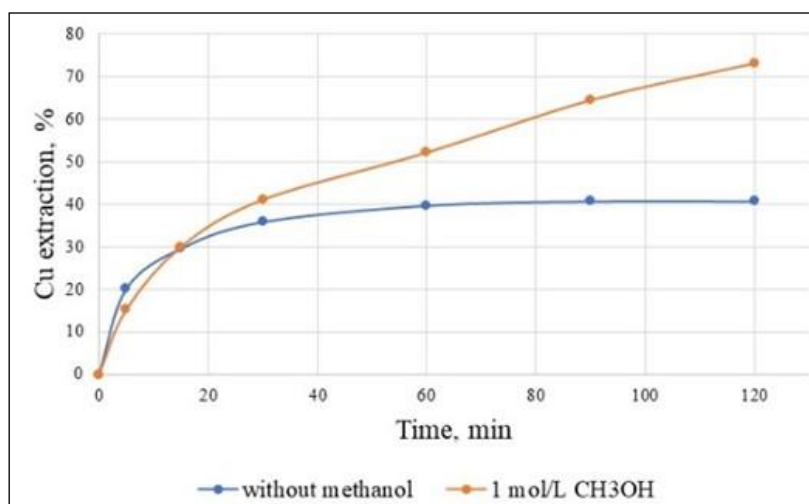


Figure 1 Copper extraction from chalcopyrite concentrate with and without methanol

(Conditions: $3 \text{ mol/L H}_2\text{SO}_4$, $2 \text{ mol/L H}_2\text{O}_2$, 40°C , 400 min^{-1} , 120 min) [23]

In the presence of 1 mol/dm^3 methanol, copper leaching degree increased more than 30 % compared to the extraction value obtained in the acidic peroxide solution without methanol. The results in Fig. 1 show that copper extraction reaches 73 % after 120 minutes in the presence of methanol. In the $\text{H}_2\text{SO}_4\text{-H}_2\text{O}_2$ solution, in the absence of methanol, the oxidation reaction of chalcopyrite essentially ceased after 30 minutes and the plateau appears on the dissolution curves indicating that the oxidation practically ends. The improvement in chalcopyrite dissolution with polar organic solvent may be attributed to the stabilization of hydrogen peroxide.

Organic reagents used to leach chalcopyrite, along with references are shown in Table 1.

Table 1 Chalcopyrite leaching studies in the presence of organic reagents

Chalcopyrite sample	Leaching solution	Reference
concentrate	$\text{H}_2\text{SO}_4\text{-H}_2\text{O}_2\text{-Acetone}$	[6]
	$\text{H}_2\text{SO}_4\text{-H}_2\text{O}_2\text{-Ethylene glycol}$	
	$\text{H}_2\text{SO}_4\text{-CuSO}_4\text{-Acetone}$	
pure mineral and concentrate	$\text{H}_2\text{SO}_4\text{-CuSO}_4\text{-O}_3\text{-Formic/Acetic acid}$	[16]
	$\text{H}_2\text{SO}_4\text{-CuSO}_4\text{-O}_3\text{-Methanol/Ethanol}$	
relatively pure chalcopyrite concentrate	$\text{H}_2\text{SO}_4\text{-H}_2\text{O}_2\text{/(CuSO}_4\text{-O}_3)\text{-Methanol/2-propanol}$	[17]
concentrate	$\text{H}_2\text{SO}_4\text{-H}_2\text{O}_2\text{-Ethylene glycol}$	[18]
concentrate	$\text{H}_2\text{SO}_4\text{-H}_2\text{O}_2\text{-Acetone/Ethanol/Ethylene glycol/Isopropanol/Methanol}$	[19]
chalcopyrite mineral	$\text{H}_2\text{SO}_4\text{-H}_2\text{O}_2\text{-Ethylene glycol}$	[22]

Conclusion

Chalcopyrite presents the most important mineral of copper, but it is a refractory mineral and its dissolution in various leaching solutions is very difficult. The results obtained in numerous leaching studies of chalcopyrite leaching with hydrogen peroxide as an oxidizing agent have shown that the presence of polar organic solvents in the leach liquor increases the copper extraction from chalcopyrite, probably due to the stabilization of hydrogen peroxide. Furthermore, the used organic additives in the chalcopyrite leaching process could modify the porosity as well as the thickness of the products layer formed during the oxidation responsible for the passivation phenomenon.

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