

Effect of initial pH on chalcopyrite oxidation dissolution in the presence of extreme thermophile *Acidianus manzaensis*



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Received 21 June 2013; accepted 5 November 2013

Abstract: The influence of initial pH on the chalcopyrite oxidation dissolution at 65 °C was investigated by bioleaching and cyclic voltammetry experiments, and the oxidation products were investigated by XRD and Raman spectroscopy. Bioleaching results show that chalcopyrite dissolution rate increases with the decrease of the initial pH in chemical leaching, while the influence of initial pH on bioleaching is on the contrary. The presence of *Acidianus manzaensis* does not promote chalcopyrite dissolution under initial pH 1.0, which mainly results from serious inhibition of high acidity to the growth of *Acidianus manzaensis*. Electrochemical experiments results show that anodic oxidation currents of electrolyte with or without *Acidianus manzaensis* both increase with the increase of initial pH, and covellite and sulfur are detected on the electrode surface. The results confirm that chalcopyrite dissolution in chemical leaching is under the combined action of oxidation and non-oxidation of proton, with conversion of chalcopyrite to covellite and elemental sulfur.

Key words: chalcopyrite; bioleaching; *Acidianus manzaensis*; initial pH; oxidation dissolution

1 Introduction

Bioleaching technology has been successfully applied to the extraction of copper from low-grade secondary copper sulfide chalcocite. However, bioleaching of chalcopyrite, the most abundant primary copper sulfide, has not been successful on a commercial scale, which is mainly due to the low leaching kinetics and copper recovery by typical mesophiles [1]. Bioleaching of chalcopyrite with thermophiles may effectively promote the dissolution of chalcopyrite [2].

It is well known that chalcopyrite is acid-soluble, and it dissolves under the oxidation action of Fe^{3+} and proton attack via polysulfide mechanism. Therefore, pH is an important influence factor for chalcopyrite bioleaching. Some researchers reported that chalcopyrite leaching rate increases with the increase of acidity in chemical leaching [3,4]. While, CORDOBA et al [5] and ANTONIJEVIĆ and BOGDANOVIĆ [6] found that high

acidity hampers the dissolution of chalcopyrite in chemical leaching. The influence of pH in chalcopyrite bioleaching is more complex than in chemical leaching, which affects not only the dissolution of chalcopyrite, but also the oxidation activity and growth of microorganisms.

Chalcopyrite (bio) leaching can be looked upon as electrochemical anodic oxidation and cathodic reduction reactions, thus it can be studied by electrochemical methods. It was reported that chalcopyrite anodic oxidation currents increase with the increase of pH [7–9]. However, ZHU et al [10] suggested that the current of the redox reaction increases with the decrease of pH. The influence of initial pH on chalcopyrite bioleaching and anodic oxidation is still unclear.

In the present study, the influence of initial pH on chalcopyrite oxidation dissolution at 65 °C was investigated by (bio) leaching and cyclic voltammetry experiments, and the solid products formed during bioleaching and on the electrode surface were studied by

XRD and Raman spectroscopy, respectively.

2 Experimental

2.1 Chalcopyrite concentrate

Chalcopyrite concentrate used in this study was the same as that by HE et al [11], and the particle size of sample was less than 75 μm .

2.2 Strain and culture medium

The strain *A. manzaensis* YN-25 used in this study was isolated by the Key Laboratory of Biometallurgy of Ministry of Education of China, Changsha, and the characteristics of the strain were described by DING et al [12]. *A. manzaensis* YN-25 was cultured in 9K basal medium supplemented with 0.02% (w/v) yeast extract, and it was gradually adapted to 0.5% (w/v) chalcopyrite for several months before using in bioleaching experiments. The cells were harvested by filtration (through 5 μm filter membrane) and centrifugation (10000 r/min) for 30 min, and the cell pellets were washed twice with sulfuric acid (0.1 mol/L) and suspended in sterilized fresh medium.

2.3 Leaching experiments

All leaching experiments were carried out in 250 mL Erlenmeyer flasks containing 100 mL sterilized medium at the pulp density of 2%. Chemical leaching experiments were conducted at three pH values (1.0, 1.5 and 2.0). The conditions of bioleaching were taken as those of chemical leaching except *A. manzaensis* and 0.02% (w/v) yeast extract was provided. The initial cell concentration in all the bioleaching experiments was $1 \times 10^7 \text{ mL}^{-1}$. The cultures were incubated at a high-temperature water bath rotary shaker at 125 r/min and 65 $^{\circ}\text{C}$. Triplicate experiments were performed under identical conditions. The water evaporation and the loss due to sampling for analyses were compensated with sterilized fresh medium.

Cell number was determined by blood cell counting chamber. The concentrations of total iron and copper ions were determined by atomic absorption spectrophotometry (AAS), and the concentrations of ferrous ions were determined by titration with potassium dichromate. The pH values of leaching solution were measured with pH meter (PHS-3C). Redox potential was measured with Pt electrode, using calomel electrode ($\text{Hg}/\text{Hg}_2\text{Cl}_2$) as reference. The preparation of samples for XRD analysis was performed as that by XIA et al [13].

2.4 Electrochemical experiments

The preparation of electrode, electrolyte, reagents, electrochemical cell and electrochemical workstation were the same as those in our previous work [14]. The

9K basal medium was used as the electrolyte and its pH was adjusted to conducted values with 1 mol/L sulfuric acid before electrochemical experiments. The temperature of electrochemical measurement was maintained at 65 $^{\circ}\text{C}$ with a thermostat.

Open circuit potential (OCP) of the massive chalcopyrite electrodes was measured, which was 0.422 V (vs SHE) after reaching its steady state for 30 s. Cyclic voltammetry experiment was performed at a potential sweep rate of 20 mV/s, scan was initiated from the OCP in positive direction, without stirring the electrolyte solution. All potentials in this work were measured (vs SHE).

Potentiostatic oxidation modification of the electrode at 0.607 V for 4 min, and preparation of the electrode sample for Raman spectroscopy analysis were performed as those in our previous work [14].

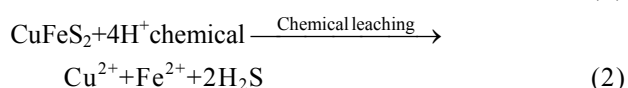
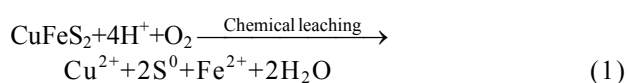
2.5 Mineral morphology and composition analyses

XRD and Raman spectroscopy analyses of the anodic oxidation products formed on the massive chalcopyrite electrode surface were performed as those by XIA et al [13]. Microscopic structure of chalcopyrite massive electrode was analyzed by Leica DMRXE automatic image analyzer (German).

3 Results and discussion

3.1 Influence of pH on chalcopyrite bioleaching

The characteristics of chalcopyrite chemical leaching under different initial pH (1.0, 1.5 and 2.0) are shown in Fig. 1. It can be seen from Figs. 1(a), (c) and (e) that the copper ion concentration reached 2.12, 0.26 and 0.23 g/L after 10 d leaching under the initial pH of 1.0, 1.5 and 2.0, respectively. The decrease of the initial pH from 2.0 to 1.0 increased the copper ion concentration from 0.23 to 2.12 g/L. The results were in accordance with those by VILCÁEZ et al [4]. Chalcopyrite dissolution rate increases with the decrease of the initial pH for the dissolution of chalcopyrite in chemical leaching, which could be mainly under proton attack as shown in the following reaction (1). Some researchers also suggested that there might be a parallel, non-oxidative dissolution process (reaction 2) [15]. The dissolution of chalcopyrite in chemical leaching could thus be under the combined action of oxidative and non-oxidative mechanisms.



As shown in Fig. 1, pH values in chemical leaching were all kept increasing for the dissolution of

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