

Comparative study on the bioleaching, biosorption and passivation of copper sulfide minerals



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ABSTRACT

The bioleaching, biosorption and passivation of five different types of copper sulfide minerals (djurleite, bornite, covellite, pyritic chalcopyrite, and porphyry chalcopyrite) by *Acidithiobacillus ferrooxidans* (*At.f* LD-1) were investigated in this paper. The bioleaching experiments showed that copper extraction followed the order djurleite > bornite > pyritic chalcopyrite > covellite > porphyry chalcopyrite. The bacterial adsorption experiments indicated that the bacterial adsorption quantity onto the five copper sulfide minerals had the same order as that of copper extraction. SEM analysis showed that bacterial distribution on the djurleite, bornite, and covellite was uniform and that on the pyritic chalcopyrite and porphyry chalcopyrite was uneven (distributed as continuous chain structure). Zeta potential analysis showed that the zeta potentials of copper sulfide minerals after interaction with *At.f* LD-1 cells changed to varying degrees. The variation degree of the zeta potentials of the five copper sulfide minerals showed the same order as the bacterial adsorption quantity on the five copper sulfide minerals. The main passivation layers on the residues of djurleite and bornite were both jarosite, and that the passivation layers on the residues of covellite, pyritic chalcopyrite, and porphyry chalcopyrite were Cu_4S_{11} , S_8 , and $\text{Cu}_4\text{Fe}_2\text{S}_9$, respectively. The inhibition ability of passivation layers followed the order $\text{Cu}_4\text{Fe}_2\text{S}_9 > \text{Cu}_4\text{S}_{11} > \text{S}_8 > \text{jarosite}$.

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1. Introduction

Biohydrometallurgy can be used to process low-grade copper minerals and refractory ores efficiently because of its low production costs and minimal environmental pollution (Gomes and Frenay, 1996; Ehrlich, 2001; Fu et al., 2008; Vakylabad et al., 2011). Recently, researchers have investigated the bioleaching of various types of sulfide ores from different perspectives. Dew et al. studied the bioleaching of various types of sulfide minerals and found that their leaching rates follow the order chalcocite > bornite > cubanite > covellite > pyrite > enargite > carrollite > chalcopyrite (Dew et al., 2000). Wu et al. studied the thermodynamic properties of various types of sulfide minerals and found that the leaching rates of different copper sulfides under direct bacterial effect follow the order chalcopyrite > copper > covellite > enargite > chalcocite > bornite. They also found that the leaching rate order under indirect bacterial effect

was copper > enargite > chalcopyrite > chalcocite > covellite > bornite (Wu et al., 2005). Yang et al. studied the bioleaching of Minyue and Dahongshan copper mines which contain mainly chalcocite and chalcopyrite, respectively. They found that the bioleaching effect of Minyue copper mine was better than that of Dahongshan copper mine. However, studies focused on determining the reasons for the leaching difference among different types of copper sulfides are limited (Norris et al., 2010).

The leaching process and leaching efficiency are affected by many factors and biosorption on the mineral surface is one of the most important among them (Rawlings, 1996; Porro et al., 1997; Behera et al., 2011). Bacterial adhesion onto minerals is the first step in most bioleaching processes. The amount of adsorbed bacteria and the adsorption rate may be directly related to the corresponding metal dissolution rates. The adhesion of *Acidithiobacillus ferrooxidans*, which have been used for bioleaching various minerals (Ohmura et al., 1993; Lors et al., 2009), onto mineral surfaces significantly, affects the bioleaching of minerals (Rohwerder et al., 2003; Zhu et al., 2012). Most bacteria are attracted to mineral surfaces by chemotaxis (White et al., 1995; Rodríguez et al., 2003), and bacterial cells often adsorb onto the crystal boundaries of target minerals (Sanhueza et al., 1999). Bacterial adhesion depends on both the biochemical properties of the bacteria and the

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interfacial properties of the minerals in a bioleaching system (Mao et al., 1994; Xia et al., 2008). Bacterial-mineral interactions can change the surface properties of minerals in association with bio-oxidation mechanisms (Chen et al., 2008; Devasia and Natarajan, 2010). Passivation is another most important factor influencing the leaching rate (Haekl et al., 1995). A dense passivation film is formed on mineral surfaces as the leaching reaction proceeds in the copper sulfides bioleaching process. This passivation layer can hinder the transfer of metal ions, O_2 , CO_2 and nutrients between the mineral surface and leaching solution, and thus inhibits the further leaching of copper (Córdoba et al., 2009). Sulfur film and jarosite are reportedly the main passivation films in the bioleaching process of copper sulfides (Shu et al., 2006). Generally, sulfur film is the main passivation film in copper sulfides leaching by mesophilic bacteria. For copper sulfide leaching by moderately thermophilic bacteria, the main passivation film is jarosite (Viramontes-Gamboa et al., 2010), because it can rapidly form at high temperatures.

This study aimed to comparatively study the bioleaching, bio-sorption and passivation of copper sulfide minerals. The amounts of adsorbed bacterial cells and copper extractions were determined. The different adsorption behaviors of bacterial cells on the copper sulfides were revealed by scanning electron microscope (SEM) and zeta potential analysis.

2. Materials and methods

2.1. Leaching bacteria

At.f LD-1, isolated from the acid mine water of a copper mine in Daye, Hubei province, China, was obtained through various stages of domestication and has good bio-activity and bio-leaching capability (Dong et al., 2011).

2.2. Copper sulfides

Five types of copper sulfides (djurleite, bornite, covellite, pyritic chalcopyrite, and porphyry chalcopyrite) were used. Djurleite (71.94% Cu, 1.22% Fe, and 20.53% S) was obtained from the Bofang copper mine in Hunan Province. Bornite (50.17% Cu, 10.67% Fe, and 24.38% S) was obtained from the Dongxiang copper mine in Jiangxi Province. Covellite (60.26% Cu, 3.20% Fe, and 33.52% S) was obtained from the Zijinshan copper mine in Fujian Province. Pyritic chalcopyrite (27.38% Cu, 28.35% Fe, and 33.31% S) was obtained from the Lizhu iron mine in Zhejiang Province. Porphyry chalcopyrite (27.88% Cu, 28.29% Fe, and 32.36% S) was obtained from the Dexing copper mine in Jiangxi Province. The purities of these five copper sulfides were 90.65%, 79.22%, 90.48%, 79.22%, and 80.62%, respectively. The particle sizes of mineral samples were all less than 74 μm . Their mean particle sizes were 47.32, 37.13, 23.92, 50.55 and 27.76 μm , respectively, and their specific surface areas were 302.67, 818.93, 1319.34, 754 and 1362.5 cm^2/g , respectively.

2.3. Microorganism culture

The culture medium was improved 9K medium, consisting of the following compounds (in g L^{-1}): $(\text{NH}_4)_2\text{SO}_4$ 2.0, K_2HPO_4 0.5, $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ 0.5, KCl 0.1, and $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ 44.2. The optimal cultivation conditions of *At.f* LD-1 were 30 °C, pH of 2.0, and rotation speed of 160 rpm (Dong et al., 2011).

2.4. Bioleaching experiment

Bioleaching experiments were performed in 250 mL flasks containing 90 mL of sulfuric acid solution (pH 2) and 10 mL of inocula. The initial cell density of the inoculated solution was

1×10^8 cells mL^{-1} . The mineral concentration was 2% (wt/vol). The flasks were kept at 30 °C and shaken at 160 rpm. The copper ion concentration in the leaching solution was determined at 4 day intervals.

2.5. Bio-sorption experiment

0.5 g of copper sulfides powder was added to flasks containing 50 mL of bacterial suspension (pH 2) with a concentration of 4.0×10^8 cells mL^{-1} . The *At.f* LD-1 bacterial solution and minerals were allowed to interact under the suspension conditions using a magnetic stirrer at 200 rpm and 30 °C. Then, centrifugation was performed for 10 min at 1000 rpm to separate the minerals and supernatant (Jia et al., 2007). Finally, the numbers of *At.f* LD-1 bacteria in the supernatant were counted under a microscope at regular 10 min intervals. By comparing the cell density in the original bacterial solution with those in the supernatant and flushing fluid, the adsorption rates of *At.f* LD-1 on the surfaces of copper sulfide minerals were obtained. The mineral samples after interaction with *At.f* LD-1 cells were then freeze dried and used for SEM analysis.

2.6. Analytical procedure

The concentration of dissolved copper ions in the leaching solution was analysed by atomic absorption spectrometry. Ferrous iron was determined by titration with potassium dichromate (K_2CrO_7). The pH and redox potential were measured using an S20 SevenEasy pH/Eh process controller. The bacterial numbers were determined by blood cell counting chambers under a ZBM-300E biological microscope. The particle size distribution and specific surface areas of copper sulfide were tested by a Winner2000 laser particle size analyser. The adsorption characteristics of *At.f* LD-1 on the mineral surface were observed by SEM analysis using a JSM-6510A scanning electron microscope. The zeta-potential of the bacterial cells was measured using a Brookhaven Zeta PALS Ver.5.57 type electro-kinetic instrument in a KCl solution with the ionic strength of 0.001 mol/L and the initial cell concentration of 4×10^8 cells/mL.

3. Results and discussion

3.1. Bioleaching of different copper sulfides

The optimal initial cell density of the inoculated solution and pulp density of the five copper sulfides were obtained from our

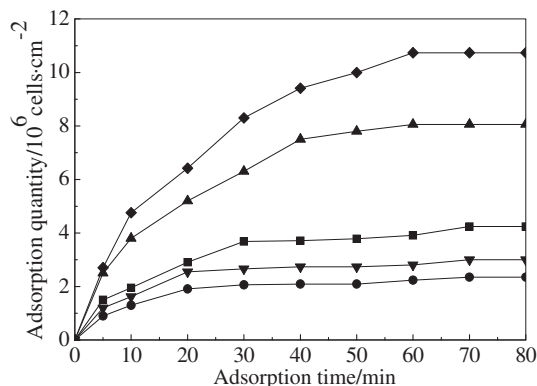


Fig. 1. Bacterial adsorption quantity on different copper sulfides vs. the adsorption time (◆: djurleite; ▲: bornite; ▼: covellite; ■: pyritic chalcopyrite; ●: porphyry chalcopyrite).

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