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# Leaching of malachite using 5-sulfosalicylic acid

Jiushuai Deng<sup>a</sup>, Shuming Wen<sup>a</sup>, Qiong Yin<sup>b</sup>, Dandan Wu<sup>a,\*</sup>, Quanwei Sun<sup>a</sup>

<sup>a</sup> State Key Laboratory of Complex Nonferrous Metal Resources Clean Utilization, Faculty of Land Resource Engineering, Kunming University of Science and Technology, Kunming 650093, PR China

<sup>b</sup> Faculty of Ming, Kunming Metallurgy College, Kunming 650033, PR China

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#### ABSTRACT

In this study, an attractive organic complexation reaction reagent with copper ions was developed for copper leaching from copper oxide ores. The possible chemical reactions of 5-sulfosalicylic acid (5-SSA) in the malachite aqueous solution were determined. Moreover, the influences of various factors, such as time, stirring speed, temperature, reagent concentration and particle size, on the leaching of copper oxide ore were investigated to establish the reaction kinetics. Results show that 5-SSA not only provided an acidic medium but also served as a complexation reagent with copper ions in the solution; thus, 5-SSA exhibited a novel leaching effect on copper oxide ores. The influence of temperature, reagent concentration and ore particle size on the leaching rate of copper was significant. The reaction kinetic equation associated with these major factors was constructed as  $1-(1-x)^{-1/3}=[0.0021 (C)^{0.638} (P)^{0.765} exp(-5103T)] t$ , with the reaction activation energy of 42.43 kJ/mol. At the same time, the stirring speed exhibited nearly no effect on the leaching rate of copper , and the dissolution process was controlled by chemical reaction. 5-SSA is therefore a new attractive reagent for copper extraction and can be extended to the metal recovery of other metal oxide ores.

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#### 1. Introduction

Copper, as one of the most common and the most valuable metals, has been extensively applied to industrial fields and every aspects of life [2,21,33,35]. In nature, ores used for copper recovery could be principally divided into copper sulfide minerals and oxide minerals, wherein chalcopyrite (CuFeS<sub>2</sub>), bornite (Cu<sub>5</sub>FeS<sub>4</sub>), covellite (CuS) and chalcocite (Cu<sub>2</sub>S), etc. are included in the former [22,25], and malachite  $(Cu_2(OH)_2CO_3)$ , chrysocolla  $[(Cu,Al)_2H_2Si_2O_5(OH)_4 \cdot nH_2O]$  and azurite  $[Cu_3(OH)_2(CO_3)_2]$ , etc. in the latter [10,26]. The copper sulfide minerals have better floatability, from which copper concentrates could be easily obtained by flotation. Nevertheless, the high-grade copper sulfide mineral resources have tapered, consequently, copper oxide ores become significant resources for copper recovery [25]. Additionally, they are widely distributed, with a large quantity included in many copper ore deposits [18]. It has become an important research point to extract copper from copper oxide ores containing carbonates and silicates [1,14,24,29]. Relative to copper sulfide ores, copper oxide ores are difficult to be separated by flotation, but they have larger solubility. As a consequence, hydrometallurgy becomes a favorable approach for copper recovery from copper oxide ores [5].

\* Corresponding author. Fax: +86 8715187068. *E-mail address:* wdd1006530@sina.com (D. Wu). With certain reagents added into the ore pulp solution, the copper oxide minerals could be dissolved, subsequently, copper could be eventually gained through extraction and electrowinning. At present, inorganic acid reagents, such as sulfuric acid, are mainly employed [4,7,15,20]. Application of organic acid as the leaching reagent also attracts researchers' interest [5,17,34].

In addition to acid leaching, ammonia leaching has presently achieved extensive applications, with the principle that complexation reaction takes place between the copper ions and the ammonia to form stable complex compound, which can be represented by the following reaction equations [3]:

$$Cu^{2+} + 2NH_3 \leftrightarrows Cu(NH_3)_2^{2+} \tag{1}$$

$$Cu(NH_3)_2^{2+} + 2NH_3 \leftrightarrows Cu(NH_3)_4^{2+}$$
(2)

Ammonia leaching of copper oxide ore has become a developed technique with novel extraction effect. However, copper extraction through complexation reaction are reported only limited to ammonium hydroxide or ammonium salts, such as ammonium chloride, ammonium sulfate and ammonium nitrate, *etc.* [3,8,9,11,19,23,31], while other complexation reactions have been rarely reported till now.

5-SSA, a derivative of salicylic acid, serves as a significantly important reagent in the biological and pharmaceutical industries. 5-SSA is easy to form ligand with Cu(II) ions [30], and potential

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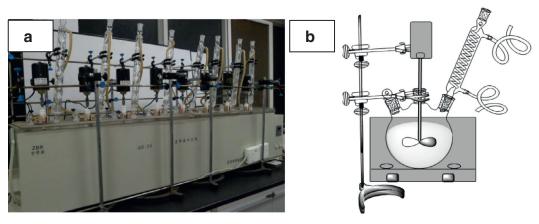


Fig. 1. Compound device for leaching (CDL), (a) Overall; (b) Structural unit.

complexation reaction could occur in the aqueous solution. Furthermore, the functional groups contained in 5-SSA could be partially or completely deprotonated to get the aqueous solution acidic [12], so that the copper oxide ore could be dissolved in the acidic environment.

Based on its ionization and complexation properties, 5-SSA can be employed as a potential leaching reagent for oxide ores. But so far, in addition to acid leaching and ammonium leaching, the application of 5-SSA in the leaching of metal oxide ores has never been investigated. In this paper, it was employed in the leaching of copper oxide ore for the first time, wherein its leaching conditions were determined; meanwhile, the influences of temperature, reagent concentration, ore particle size and stirring speed on the leaching rate were thoroughly studied, and the leaching kinetics was illustrated through the shrinking core model, thus the overall experimental parameters comprehensively reflecting 5-SSA as a new potential leaching reagent were eventually obtained, which could also provide a reference for the application of such reagent in the metal recovery from other metal oxide ores.

### 2. Materials and methods

#### 2.1. Materials

The experimental materials were taken from the copper mining area in northeast Yunnan in China. Firstly, ores with highergrade copper were picked out, which were subsequently crushed and screened to remove the gangues such as quartz and calcite, *etc.* as much as possible, and finally, ore sample with an average particle size of 2000  $\mu$ m was obtained.

In this experiment, de-ionized water was used to prepare the leaching solvent, which was prepared by Mill-Q5O water-purifying device made in America, with the resistivity of 18.25MQ; additionally, the 5-SSA used here was analytically pure, produced by FUYU Fine Chemical Engineering Co., Ltd. in Tianjin, China. A solution of 5-SSA was the leaching solvent.

#### 2.2. Procedure

A suit of compound device for leaching (CDL) was particularly self-designed, as shown in Fig. 1. Therein, 8 reactor units were all placed into a thermostatic water bath, with each connected by one three-necked flask, and for each flask, a Graham condenser and a mechanical stirrer were placed in its two necks, respectively, and the third one was employed for sample filling and pipetting, which was blocked off by a ground stopper. In the CDL, the 8 Graham condensers were connected end to end by rubber tubes, and respectively connected to the water inlet and outlet, so as to provide condensation for the steam generated during the experiment. Thus, the whole CDL was constituted of one water bath, eight three-necked flasks, eight mechanical stirrers and eight condensers, wherein a number of magnetic stirrers were placed at the bottom of the water bath, which would continuously stir to get the liquid homogeneously mixed. Moreover, the water bath temperature could be automatically controlled, and the rotating speed of the mechanical stirrers could be adjusted. As a consequence, eight groups of experiments could be simultaneously conducted by the CDL, thus both the consistency of the experimental conditions and the accuracy of the experimental results could be easily guaranteed.

After the solution temperature in the water bath was well regulated, 5 g ore sample and 1000 ml 5-SSA solution were respectively added into each three-neck flasks to conduct the leaching experiment, wherein the effects of temperature, reagent concentration, stirring speed and particle size, *etc.* on the 5-SSA leaching malachite were focused to be researched. During the process, 5 ml solution was pipetted by a pipette and filled into a sealed chemical bottle for each time, and next, inductively coupled plasma atomic emission spectroscopy (ICP-AES, ICP-1000 II SHIMADZU) was applied to test the copper concentration in the solution at a certain moment, and furthermore, the leaching rate *x* was calculated.

## 3. Results and discussion

#### 3.1. Characterization of the samples

So as to test the composition of the samples, X-ray diffraction analysis (XRD), chemical titration method, scanning electron microscope (SEM) and energy dispersive spectrometer (EDS) were conducted.

The model of the XRD tester was NSK D/max, with the CuK $\alpha$  target ( $\lambda = 1.5406$  Å) adopted, the voltage, the current and the scanning range of 40 kV, 40 mA and 10°–90°, respectively. The XRD pattern was shown in Fig. 2, as could be seen, there were multiple obvious peaks corresponding to the malachite crystals, whose positions well coincided with those in the Ref. [27]; moreover, there were also a small amount of quartz and calcite contained in addition to the malachite. Totally, from the XRD result, malachite was demonstrated to be the major copper mineral in the experimental sample.

The chemical titration method result was shown in Table 1, which indicated that CuO and  $SiO_2$  accounted for 57.30% and 8.97%, respectively; additionally, there were also gangue minerals containing Ca, Mg and Al as well, therefore, this copper oxide ore was principally composed of malachite.

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