ISSN 1062-7391, Journal of Mining Science, 2016, Vol. 52, No. 3, pp. 569–575. © Pleiades Publishing, Ltd., 2016.

MINERAL DRESSING

A Statistical Approach to the Experimental of the Leaching of Sulfide Copper from the Ores Using Lixiviant Sulfuric Acid[1]

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Received January 17, 2016

Abstract—In the present paper, a review has been made for the recovery of copper from the ores using lixiviant sulphuric acid, the effects of the sulfuric acid concentration, leaching time, particle size and leaching temperature on the recovery of copper were examined. The results of leaching indicate that the recovery extraction of Cu increases with increasing acid concentration and temperature; while it decreases with particle size. The logical experimentation parameters for the extraction of copper were discovered. Later than 120 min of leaching treatment, the sulfuric acid concentration 2 mol/l^{-1}, leaching temperature 85 °C, smaller particle size 63 µm at low stirring rates (100 rpm) and solid-to-liquid ratio 1:10. The overall results of the dissolution studies indicated that the data fitted the shrinking core model for the controlled mechanism, with surface chemical reaction as the rate controlling step. The value of the activation energy Ea is calculated by multiplying the slope of the Arrhenius curve by the value of the universal gas constant 8.314 kJ/mol^{-1}.

Keywords: Copper sulfide concentrate, leaching, sulfuric acid, dissolution kinetics.

DOI: 10.1134/S1062739116030849

INTRODUCTION

Leaching is an interaction process between dissolved reagents and solid phase. Temperature, concentration of reagents, rate of mixing, surface area of solid phase and other parameters affect the rate of leaching [1]. The shrinking core model consider as the leaching process is controlled either by the diffusion of reactant through the solution boundary, or through a solid product layer, or by the surface chemical reaction rate. Assuming that samples particles have a spherical geometry and surface chemical reaction is the slowest step [2].

Pyrite and chalcopyrite, the most common and exploitable sulphide minerals, usually occur together and in contact with each other [3]. Copper sulfides describe a family of chemical compounds and minerals. Prominent copper sulfide minerals include CuS (covellite) and chalcopyrite, which consist of mixed copper-iron sulfides. It is generally coupled with supplementary sulphide minerals, such as chalcopyrite ($CuFeS_2$), galena (PbS), pyrite (FeS_2) and tedrahedhrite $(Cu, Fe)_{12} (Sb, As)_4S_{13}$ [4–8].

The leaching of copper is more favorite since it’s minor energy cost; low speculation costs; the possibility of treating ores in situ (in the case of heap leaching); and the continuous extraction processes. Most copper ores are now extracted metal sulfides such as chalcopyrite. One solution is simply to liberate Cu^{2+} ions by spraying a gang of ore with sulfuric acid. The majority common reagent used in leaching of metals is sulphuric acid (H_2SO_4). Whereas other heavy metals are talented to dissolve in H_2SO_4, copper (Cu) dissolves quicker in this acid than metals such as zinc, cobalt and iron because it is higher up in the electrochemical series.

The study kinetics of leaching is very significant to understand the dissolution process of metals. Lochmann show that shrinking core model is classical model use for illustration leaching mechanism of sulphide minerals [9]. Many authors like supported the shrinking core model in sulphides ores leaching [10–12].

In this paper, a review has been made for the recovery of copper from the ores using lixiviant

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[1] The article is published in the original.

sulphuric acid, to determine the most favorable temperature leaching, particle size and discovered the mechanism of the dissolution reaction.

1. EXPERIMENTAL

1.1. Material

The bulk concentrate from this ore was produced at cheabet Elhamra mine, Setif, Algeria. Table 1 show the chemical compositions of the bulk concentrate.

1.2. Leaching Procedure

The leaching experiments were carried out in flask fitted with a reflux condenser and a mechanical stirrer. The flask was placed on a thermostatically controlled heating mantle. The solution temperature was controlled to specific values with continuously monitored by a thermometer. Samples of clay weighing 10 g were taken in 100 ml of leaching solution. The order of levels was randomly chosen as the order of experiments to avoid any bias.

The optimum concentrations of the acids were determined by leaching in (0.25, 0.5, 1 and 2 mol/l^{-1}) sulfuric acid solutions. The subsequent leaching was performed in the optimized acid concentrations at temperatures of (25, 45, 65 and 85 °C) for (30, 60, 90, and 120 minute) at each temperature on low stirring rates (100 rpm). At the finish of leaching for a specific period of time at a specific temperature of leaching solution was taken out of the round bottom flask by a pipette. The collected sample of leach liquor was cooled, filtered. Metal content was analyzed by atomic absorption spectrometry (AAS Shaker D407).

2. RESULTS AND DISCUSSION

2.1. Results of Leaching

2.1.1. Effect of acid concentration

To investigate the effect of sulfuric acid concentration on the leaching of extraction copper, the leaching was studied at different experiments are on 63 μm of particle size was carried out by varying the initial sulfuric acid concentration over 0.25–2 mol.l^{-1} at 65 °C for leaching time varying between 30 and 120 min, and a solid-to-liquid ratio (1:10). The H_2SO_4 concentration has a significant effect on the leaching of extraction copper. Figure 1 show the effect of sulfuric acid concentration and leaching time on the leaching of extraction copper. As it can be seen, the leaching of extraction copper increased to 20.26 % with an increase in H_2SO_4 concentration from 0.25 to 2 mol/l^{-1} at the leaching time of 120 min. Therefore, these values can be chosen as the optimum reaction solution.

Table 1. Chemical composition of hydrometallurgical residue (mass fraction, %).

Elements	Zn	S	Fe	Cu
	41.61	39.38	13.71	0.14

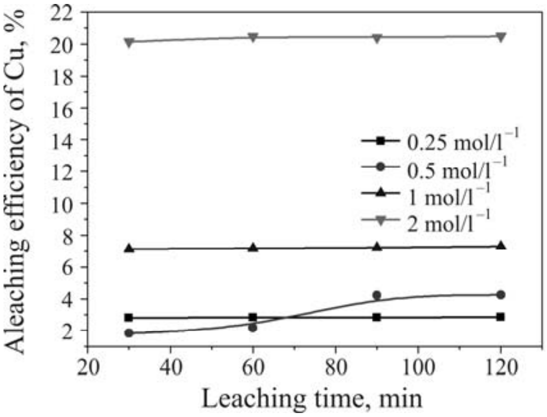

Fig. 1. Effect of H_2SO_4 concentration on copper dissolution at various leaching time. Experimental conditions: particle size = 63-125μm; temperature = 65°C.

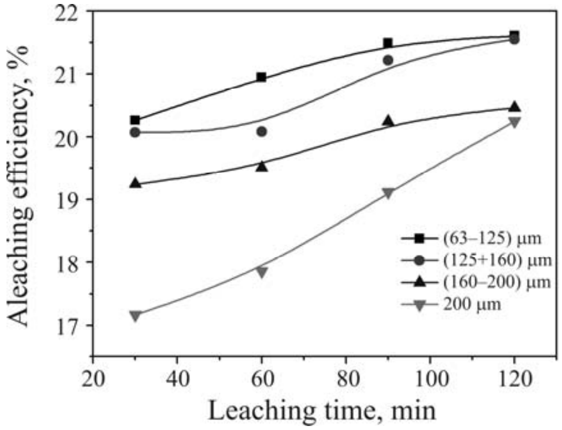

Fig. 2. Effect of leaching time at different particle size.

2.1.2. Effect of leaching time

Generally study; the leaching time is important parameter in the thermodynamic behavior, because the reaction duration is sensible to the leaching of extraction copper, the letter was studied as a function of leaching time with different particle sizes at a leaching temperature of 85 °C with sulfuric acid concentration of 2 mol/l^{-1}, where the solid-to-liquid ratio of (1:10). Figure 2 demonstrates that the solubility of sample in acidic solution is related the reaction duration. It is seen in the figure that with increasing the leaching time, the leaching rate increased in all particle sizes; but when the leaching time was over 60 min, the curve of leaching rate was changed to be steady; the suitable leaching time is 120 min, reaching the maximum extraction of copper is 20.26 % obtained with 63 μm at a leaching time of 120 min.

Experimental conditions: temperature = 85°C; [H2SO4] = 2 mol/l-1.

2.1.3. Effect of particle size

To obtain more information on the leaching of extraction copper, this letter was studied as a function of leaching temperatures at various particle size, which investigate with the constants of sulfuric acid concentration (2 mol/l^{-1}), leaching time (120 min) and solid-to-liquid ratio (1:10). Figure 3 shows the effect of the particle size on leaching process at various leaching temperatures: the maximum extraction rate of copper was obtained at the particle size of 63 μm, after this point the leaching rate was changed to minimum value. The good result was achieved in the particle size of 63 μm at 85 °C. Li et al. [14] they have observed that the particle size of the sample chosen was as 48–42 μm to obtain a higher extraction percent of zinc and indium. This is in agreement with [15, 16].

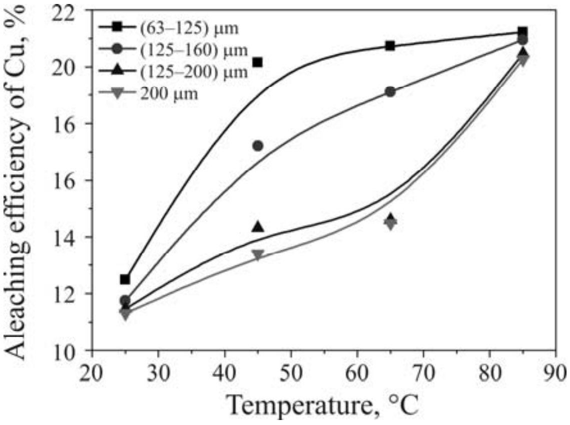

Fig. 3. Effect of particle diameter on copper dissolution in [H_2SO_4] = 2 mol/l^{-1}.

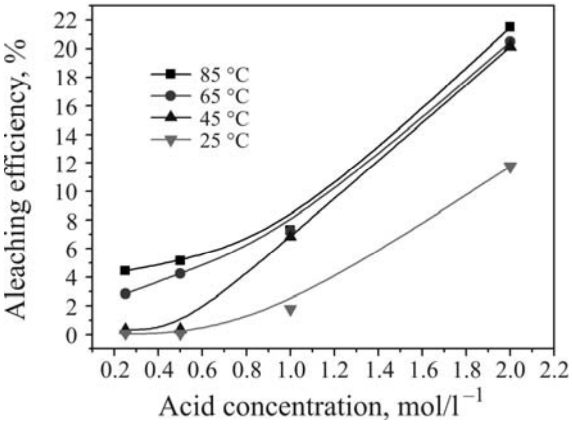

Fig. 4. Effect of temperature on leaching rate of sulfide copper.

2.1.3. Effect of leaching temperature

The leaching temperature is important parameter in the solubility of the recovery of copper dissolution in sulfuric acid, the extraction copper was studied at different sulfuric acid concentration, solid-to-liquid ratio (1:10), particle size (63 μm), at leaching times 120 min. These results are presented in Fig. 4 showing variation of the leaching rate of copper as a function of the leaching temperature. As it can be seen, also the leaching rate of copper increase with increasing the leaching temperature, reaching the maximum extraction of copper was found of 20.26%, it is obtained with sulfuric acid concentration 2 mol/l^{-1} at a leaching temperature of 85 °C, can be compare with 90 min was found of 20.91%, it is in qualitative agreement to obtain the maximum enhancement. This observation was also supported by [13–19].

Experimental conditions: particle size (63 μm), leaching time is 120 min.

2.2. Discussion of Kinetic Models

2.2.1. Copper dissolution kinetics studies

If we consider X as the fraction leached at time t, the fraction not dissolved is given by the following expression of shrinking core model can be used to describe the dissolution kinetics (Fig. 5).

$$1 - X = \left(1 - \frac{k't}{r_i}\right)^3, \tag{1}$$

$$1 - (1 - X)^{\frac{1}{3}} = \left(\frac{k'}{r_i}\right)t, \tag{2}$$

where $1 - X$ is the fraction no dissolved; r_i is the particle radius; k' is the constant linear velocity;

$$1 - (1 - X)^{\frac{1}{3}} = kt. \tag{3}$$

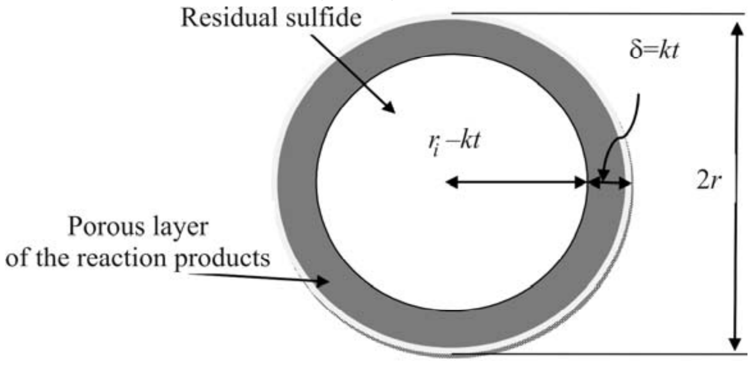

Fig. 5. Simplified shrinking core model for dissolution of copper sulfide.

Figures 6 and 7 shows that there is linearity between the term $1-(1-X)^{\frac{1}{3}}$ and leaching time, (this lines intermediate between points: are analyzed with fit linear in logiciel OriginePro 8); therefore, we can assume that the shrinking core model is consistent with the dissolution of copper concentrate processed. The modeling equations are in the type:

$$y = a + b, \tag{3}$$

where b is mainly related to the rapid dissolution of copper oxides contains in the rock unlike that of sulfides does not leave insoluble reaction products; justified that X is not zero on first emergence of reaction. This agrees with the results obtained in [20].

The experimental rate constant k was calculated from the slope of the straight line at various temperatures (Fig. 7), their equivalent correlation coefficients are summarized in Table 2.

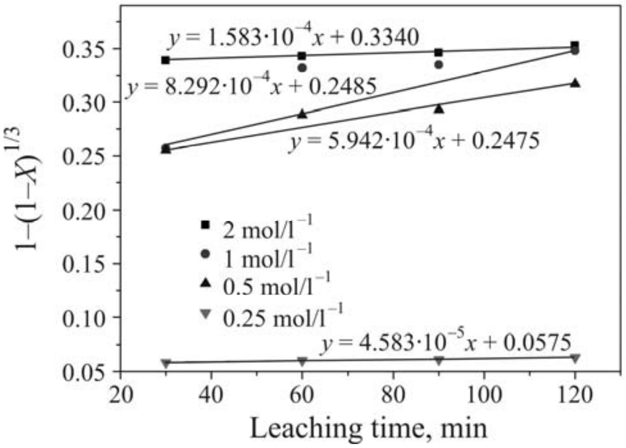

Fig. 6. Plot of 1 - (1 - X) $^{1/3}$= kt vs. leaching time at various H_2SO_4 concentrations.

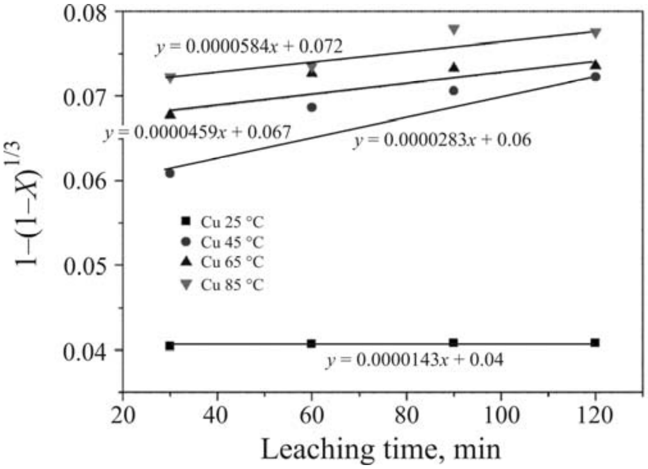

Fig. 7. Plot of 1 - (1 - X) $^{1/3}$= kt vs. leaching time at various temperature.

Table 2. The values of rate constants with their correlation coefficients for copper dissolution at different temperatures by [H_2SO_4] = 2 mol/l^{-1}.

Temperature, °C	Cu	
	K, 10^{-3}min^{-1}	R^2
25	0.1430	0.9823
45	0.0283	0.6591
65	0.0459	0.6127
85	0.0584	0.7036

2.2.2. Evaluation of activation energy of copper dissolution

The chemically controlled process is strongly dependent on temperature as demonstrated in [11]. From the slopes in Fig. 7, we use the rate constants resultant; the Arrhenius diagram in Fig. 8 was drawn via:

$$k = Ae^{-E_a/RT}, \tag{4}$$

where k is the apparent rate constant, which k is exponentially dependent on temperature; A is the Arrhenius constant; E_a is the activation energy; T is the absolute temperature; R is the universal gas constant. This law concurs to calculate the activation energy E_a, which is the minimum that the reactants must acquire to be able to react and turn into a product of leaching reaction energy.

The value of the activation energy E_a is calculated by multiplying the slope of the Arrhenius curve by the value of the universal gas constant 8.314 KJ/mol^{-1}. The calculated value is 20.9 KJ/mol^{-1} and the Arrhenius constant A is 8.27×10^{-5} min^{-1}.

CONCLUSIONS

The leaching of extraction copper from Chaabet El Hamra, Algeria bulk sulphide ore was used to recovery of copper dissolution in sulfuric acid with varying the leaching temperature, leaching time, particle size and sulfuric acid concentration. The following conclusions can be drawn from the results presented:

—the sulfuric acid dissolution of Chaabet El Hamra, Setif (Algeria) bulk complex sulphide ore was studied. The process of direct leaching of extraction copper with sulfuric acid operated at low stirring rates (100 rpm);

—the results of leaching investigations showed that the temperature is a fundamental parameter for copper extraction. The recovery of copper increases with increasing sulfuric acid concentration and decreasing particle size;

—the extraction rates from the residue after 120 min of treatment is 20.26% by the side of the optimum leaching conditions: solid-to-liquid ratio (1:10), temperature leaching 85°C, sulfuric acid concentration 2 mol/l^{-1}, and particle size 63 μm,.

—the overall results of the dissolution studies indicated that the data fitted the shrinking core model for the controlled mechanism, with surface chemical reaction as the rate controlling step. The activation energy value for copper is 20.91 KJ/mol^{-1}.

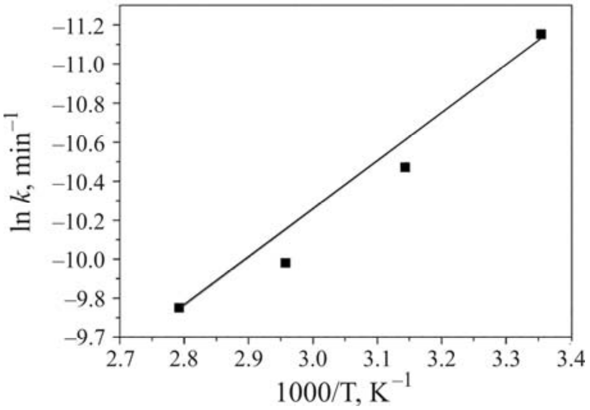

Fig. 8. Plot of ln k vs.1000/T (K-1): Arrhenius plot for determining activation energy, Ea, for sulphide copper dissolution in 2 mol/l-1 M H2SO4.

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