



Kinetics of zinc sulfide concentrate direct leaching in pilot plant scale and development of semi-empirical model



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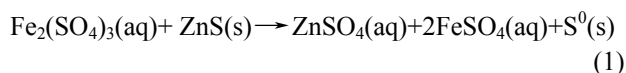
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Abstract: The direct leaching kinetics of an iron-poor zinc sulfide concentrate in the tubular reactor was examined. All tests were carried out in the pilot plant. To allow the execution of hydrostatic pressure condition, the slurry with ferrous sulfate and sulfuric acid solution was filled into a vertical tube (9 m in height) and air was blown from the bottom of the reactor. The effects of initial acid concentration, temperature, particle size, initial zinc sulfate concentration, pulp density and the concentration of Fe on the leaching kinetics were investigated. Results of the kinetic analysis indicate that direct leaching of zinc sulfide concentrate follows shrinking core model (SCM). This process was controlled by a chemical reaction with the apparent activation energy of 49.7 kJ/mol. Furthermore, a semi-empirical equation is obtained, showing that the order of the iron, sulfuric acid and zinc sulfate concentrations and particle radius are 0.982, 0.189, -0.097 and -0.992, respectively. Analysis of the unreacted and reacted sulfide particles by SEM-EDS shows that insensitive agitation in the reactor causes detachment of the sulfur layer from the particles surface in lower than 60% Zn conversion and lixiviant in the face with sphalerite particles.

Key words: kinetics; direct leaching; sphalerite; shrinking core model (SCM); pilot plant

1 Introduction

Leaching of sphalerite was, usually, carried out under pressure and oxygen plays a key role in zinc dissolution process [1,2]. Also, atmospheric leaching of sphalerite has been studied by some researchers [3–8] and it has been shown that added ferric ions or other oxidizing agents (in solution) usually oxidize sphalerite in atmospheric media. From these methods, an integrated method has, also, been proposed where oxygen gas is used to sphalerite leaching in the atmospheric media [9–11]. In this case, ferrous ionic species are oxidized by oxygen blowing in the reactor. The generated ferric ions are reduced to ferrous ion by sphalerite:



The ferrous ion (Fe^{2+}) is re-oxidized in order to continue the leaching. Therefore, concentration of ferric ions, as an oxidant, could affect kinetic of zinc sulfide leaching. Oxygen blowing rate, particle size, temperature and acid concentration are other effective factors in

direct leaching of sphalerite.

MARKUS et al [6] have investigated particle size variations during leaching process. The results confirmed the formation of the product layer on the surface particles. Moreover, other reports [1,8,12] confirmed shrinking core model (SCM) in atmospheric leaching that a sulfur layer is formed on solid particles. Three known control regimes in sulfide leaching kinetics are liquid film diffusion, solid product diffusion and chemical reaction controls. Several reports [8,13,14] have refused liquid film diffusion control, by determination of the processes activation energy (E_a). Furthermore, due to intense mixing in the reactor, sphalerite leaching may not be controlled by the liquid film diffusion.

On the other hand, a major feature of the kinetic system is the chemical reaction step and reacting mass transport coupled in series. In this case, the chemical reaction occurs in an interface between the original solid and the reaction product. If we assume that the zinc sulfide particles have a spherical geometry and the chemical reaction is the rate-controlling step, then it is expression of the shrinking core model to describe the dissolution kinetics of the process as follows:

$$1 - (1 - X)^{1/3} = k_r t,$$

$$k_r = \frac{bk_c C_A^n}{\rho(\text{ZnS})r_0} \quad (2)$$

When the diffusion of lixiviant (such as Fe^{3+}) through the elemental sulfur layer is the rate-controlling step, the following expression can be described as kinetic model:

$$1 - 3(1 - X)^{2/3} + 2(1 - X) = k_d t,$$

$$k_d = \frac{6bDC_A}{\rho(\text{ZnS})r_0^2} \quad (3)$$

where X is the fraction reacted and k_r and k_d are the apparent rate constants for chemical reaction control and product diffusion in the product, respectively. Also, k_c is the chemical reaction rate constant, C_A is the concentration of the lixiviant in the solution, b is stoichiometric coefficient of the reactant in the leaching reaction, r_0 is the initial radius of the solid particle, n is the order of reaction with respect to C_A and D is the diffusion coefficient in the product layer. If the chemical reaction on the surface or diffusion through the product layer controls the leaching rate, there must be a linear relation among the left side of Eq. (2) or Eq. (3) and time.

Results of the investigation on determination of kinetic mechanism of sphalerite leaching were totally different. DUTRIZAC [14] and PECINA et al [8] indicated that the kinetics of sphalerite dissolution in ferric sulfate media is controlled by a chemical reaction on the surface of the particles. Also, XIE et al [1] mentioned that the interface chemical reaction is the controlling step in the pressure leaching.

On the other hand, some authors [7,15] have reported that the kinetics could be controlled by a non-reaction mechanism. An investigation [16] reported that both phenomenon of diffusion in product layer and the reaction between the particle surface and the product layer play a key role in controlling reaction rate. Generally, the kinetic model of zinc sulfide leaching is affected by different factors, such as agitation rate, and minerals composition in ore [17].

Even though all these studies provide valuable background on sphalerite dissolution kinetics, their results do not cover the dissolution kinetics during direct

atmospheric leaching process. Moreover, studies about direct leaching of sphalerite [18,19] were carried out at bench scale, while the effect of various factors on direct leaching kinetics should be surveyed at the larger scale.

The aim of this work is to investigate effects of kinetic characteristics (such as kinetic control regime, activation energy of the dissolution and reaction order of different parameters) on direct leaching of zinc sulfide concentrate. All tests were performed in tubular reactor and pilot scale and then the collected data were analyzed by SCM. Influences of the particle size, acid concentration, zinc sulfate concentration, temperature, and solid to liquid (S/L) ratio were also studied on sphalerite direct saturation kinetics. The activation energy of the dissolution process was evaluated and semi-empirical rate equation was presented based on the experimental and calculated results.

2 Experimental

The zinc concentrate enriched through the flotation of Angouran mine sulfide ore in the Aria flotation plant (Zanjan, Iran) was used in this work. First, sulfide concentrate was ground in rod mill and then it was sieved to obtain the particle size distribution. Mean particle size and chemical compositions of the concentrate were determined by LPSA (laser particle size analyzer) and atomic absorption spectroscopy, respectively. The crystalline ferrous sulfate and sulfuric acid of industrial grade were used to make up all needed solutions. Analytical grade chemicals were used for solution analysis. Results of sulfide concentrate analysis can be seen in Table 1. The low content of iron is remarkable, which can influence zinc dissolution rate.

The contents of the metallic ions (Zn, Fe) in the sample solutions obtained from the leaching experiments were analyzed by atomic absorption spectroscopy (Varian, AA240). The ferrous and ferric ion concentrations were analyzed by titration of cerium (IV) sulfate with Phenanthroline indicator and EDTA with the salicylic acid indicator, respectively. The sulfuric acid concentration was determined by titration of sodium hydroxide and Methyl yellow indicator.

Leaching experiments were carried out in a tube reactor equipped with a thermometer, air inlet at bottom of tube, indirect steam coil, pump and a prime tank for

Table 1 Chemical composition of different fractions of complex concentrate

Mean diameter/ μm	w(Zn)/%	w(Fe)/%	w(Pb)/%	w(Cd)/%	w(Cu)/ 10^{-6}	w(Ni)/ 10^{-6}	w(Mn)/ 10^{-6}
52	50.8	2.6	3.6	1.65	910	463	174
65	46.5	2.1	3.3	1.6	827	433	170
85	50.6	2.4	3.1	2.1	870	450	172
115	46.7	2.5	3.46	2.1	911	715	200

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