



Leaching kinetics of antimony-bearing complex sulfides ore in hydrochloric acid solution with ozone

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Abstract: The leaching kinetics of Sb and Fe from antimony-bearing complex sulfides ore was investigated in HCl solution by oxidation–leaching with ozone. The effects of temperature, HCl concentration, stirring speed and particle size on the process were explored. It is found that the recoveries of Sb and Fe reach 86.1% and 28.8%, respectively, when the reaction conditions are 4.0 mol/L HCl, 900 r/min stirring speed at 85 °C with <0.074 mm particle size after 50 min leaching. XRD analysis indicates that no new solid product forms in the leaching residue and the leaching process can be described by shrinking core model. The leaching of Sb corresponds to diffusion-controlled model at low temperature (15–45 °C) and mixed-controlled model at high temperature (45–85 °C), and the apparent activation energies are 6.91 and 17.93 kJ/mol, respectively. The leaching of Fe corresponds to diffusion-controlled model, and the apparent activation energy is 1.99 kJ/mol. Three semi-empirical rate equations are obtained to describe the leaching process.

Key words: ozone; antimony-bearing complex sulfides ore; oxidation–leaching kinetic; mixed-controlled model; diffusion-controlled model

1 Introduction

Most antimony is usually used as flame retardants, catalysts in plastics production, pigments in paints and in production of glassware [1]. China is the largest producer of antimony in the world [2]. The most important antimony resource exploited in China is the antimony ore containing stibnite (Sb_2S_3). Traditionally, extraction of antimony from stibnite involves a two-step pyrometallurgical process including roasting and carbothermal reduction [3,4]. However, the pyrometallurgical process usually results in serious environmental pollution and high energy consumption [5].

Compared with the pyrometallurgy technology, the hydrometallurgical process is considered as an environmental friendly technique which has attracted much attention due to its potentials in dealing with low grade complex stibnite [6]. Leaching of stibnite in an alkaline solution was proposed by previous

researchers [7–10] and has been employed industrially in China, Australia and United States. In the acidic chloride system, hydrochloric acid was used as the lixiviant in conjunction with oxidants, such as ferric chloride, chlorine and antimony pentachloride for antimony recovery from stibnite [11–13].

In our previous research [14,15], the extraction of antimony from complex sulfides ore was studied and the optimized conditions were obtained. However, few research has been reported on the kinetics of antimony leaching by ozone. In this work, the kinetics of antimony and iron from complex sulfides ore and the factors affecting antimony extraction were investigated.

2 Experimental

2.1 Materials

The composition and phases of ore are reported in our previous study [14]. In the experiments, the ore was crushed to sizes of <0.074, 0.074–0.089, 0.089–0.124

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and 0.124–0.179 mm. The concentration of metal element in this ore was determined by inductively coupled plasma atomic emission spectrometry (ICP-AES, PS-6, Baird, USA) and X-ray fluorescence (XRF, S0902724, Rigaku, Japan). The mineralogical composition was identified by X-ray diffractometry (XRD, S0902240, Rigaku, Japan). At the same time, the antimony and iron in leaching solution were analyzed by ICP-AES for kinetics analysis.

Analytical grade hydrochloric acid (36%–38%, mass fraction) was used to prepare the HCl solutions. Industrial grade oxygen was used to produce ozone for the leaching process and the mass flow of the ozone controlled by an ozonizer (OZOMJB–80B, ANQIU OZOMAX EQUIPMENT, China) was 120 mg/L.

2.2 Leaching procedure

The leaching experiments were carried out in the same way as our previous study [14], except the liquid to solid ratio.

The 400 mL of HCl solution of required mole ratio diluted with water was added to the beaker. When the temperature reached the set value, 8.0 g of the antimony-bearing complex sulfides ore was added, and the ozone gas was pumped into the solution. A condenser was used to prevent evaporation during the experiment. 5.0 mL of the liquid sample was collected and tested by ICP-AES every 10 min, and at the same time, 5.0 mL of the HCl solution was added to the system to keep the solution volume unchanged. After 50 min leaching process, the slurry was withdrawn from the reactor by vacuum filtered. The residue was sent to detect after drying in oven. The volume of the filtered pregnant solution was measured by a measuring cylinder. The leaching efficiency is calculated based on the sampling solutions in pre-determined intervals and the leaching efficiency of antimony (or iron), x , is defined by

$$x = cV / (m\omega) \quad (1)$$

where c , V , m and ω are the antimony concentration in filtration, the volume (we assumed it as constant), mass and antimony (or iron) content, respectively. The effects of temperature, acid concentration, stirring speed and particle size on the leaching process were investigated.

3 Results and discussion

3.1 Leaching behavior and kinetics model of antimony

3.1.1 Effects of temperature on leaching efficiency of antimony

Figure 1 shows the effect of temperature on the leaching efficiency of antimony in the range of 15–85 °C. The operating conditions are 4.0 mol/L HCl, 900 r/min

stirring speed, 2.0 L/min gas flow rate, <0.074 mm particle size and 50 mL/g L/S ratio. The results indicate that the leaching efficiency of antimony increases with the increase of temperature and the leaching efficiency of Sb rises from 19.9% at 15 °C to 86.1% at 85 °C. This is because the mass transfer between ore, ozone gas and lixivium is promoted with the increase of temperature, and the reactions could happen more easily at high temperature.

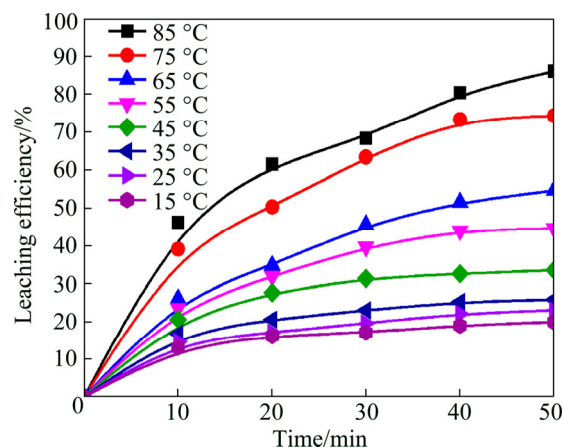


Fig. 1 Effects of temperature on leaching efficiency of antimony from antimony-bearing complex sulfides ore

According to XRD pattern of leaching residue, no sulfur was found in the leaching residue [14], meaning that the sulfur in ore was transformed into sulfate instead of elemental sulfur. There is no solid product generated in the leaching process and the leaching process can be described as typical shrinking core model [16,17]. The empirical equations are listed below:

$$1 - (1-x)^{1/3} = k_1 t \quad (2)$$

$$1 - (1-x)^{2/3} = k_2 t \quad (3)$$

$$1 - (1-x)^{1/3} + \beta [1 - (1-x)^{2/3}] = k_3 t \quad (4)$$

$$\beta = k_1 / k_2$$

where Eq. (2) shows chemical reaction control, Eq. (3) is mass-diffusion control and Eq. (4) stands for mixed-control, x stands for leaching efficiency, k_i is rate constants and t is reaction time.

In order to determine the kinetic parameters and rate-controlling step of antimony leaching process, the experimental data presented in Fig. 2 is fitted by Eqs. (2)–(4). The linear relative coefficients of plots of Eqs. (3) and (4) vs time are higher than that of Eq. (2), indicating that the diffusion control and mixed control model are suitable for the leaching process. The plots are shown in Fig. 2, where the k_2 and k_3 could be obtained. According to Arrhenius equation, it is represented as

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