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A comparison of ultrasound-augmented and conventional leaching of silver from sintering dust using acidic thiourea

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ABSTRACT

In the process of steel manufacture, up to ten millions of tons of sintering dust (SD) are produced annually in China, which contain noble metals such as Ag. Therefore, recovery of silver (Ag) from SD could be a potential economic and environmental activity. The purpose of this article is to generate information about reaction kinetics of silver leaching with thiourea from SD, comparing the conventional and ultrasonic-augment leaching. The effects of various control parameters such as the ultrasound power, particle size, leaching temperature and thiourea concentration on leaching rate of silver were studied. The results showed 89% silver recovery for conventional process against 95% for ultrasound assisted leaching. The ultrasonic wave increased the leaching rate and shorten the reaction time. The rate controlling step was analyzed using shrinking core model and the rate controlling step is identified to be the diffusion through the product layer in both conventional and ultrasonic-augment leaching processes. The activation energies were estimated to be 28.01 kJ/mol and 18.19 kJ/mol, and the reaction order were 0.89 and 0.71, respectively.

1. Introduction

Silver, one of the most important noble metal due to its unique physical and chemical properties including low melting point, best electrical conductivity, high ductility and elongation, relative chemical stability, etc [1–3], is one of the widely used metals in industry. The majority of silver is consumed in the silver halide photosensitive material, jewelry and silverware, electrical contact material and silver paste, silver composite materials, silver alloy solder, energy materials, catalyst, medical materials, and antibacterial materials, etc [4]. With the development of Chinese economy, the industrial demand of silver increased sharply, economic recovery from secondary sources become increasingly important.

Sintering dust (SD) is a kind of the dust collected by electrostatic precipitation in the iron and steel metallurgy sintering process [5]. It contains number of hazardous components and these solid waste piles are of serious environmental concern. At present, sintering dust is returned to the sintering machine to reuse Fe and C, neglecting the enrichment of some hazardous metals such as Zn, Pb, K and Na which are harmful for normal operation of blast furnace and sintering machine [6]. Statistically, approximate 4 kg of sintering dust is produced per ton steel, with the total amount being 15 million tons in 2011 [7]. In recent years, the extraction of valuable element from the sintering dusts is explored by many researchers [8–11]. However, the extraction of Ag from the sintering dusts has rarely been reported.

Over the past decades, using ultrasound-augment for ore leaching became popular in the hydrometallurgical process. Sonochemical extraction techniques was reported to increase the leaching rate, shorten the duration of leaching and improving the effectiveness in the leaching process [12,13]. Ultrasound not only can create the cavitation effect to produce many micro-cracks on the solid surface, but also increases the diffusion speed of soluble species in the liquid phase [14]. The researchers [15,16] have reported a significant increase in recovery of uranium from MgF_2 with the use of ultrasonic leaching as compared to conventional mixing methods under various acid concentrations and particle sizes. Swamy et al. [17] have designed a dual frequency ultrasonic leaching equipment for the recovery of copper from the oxide ore with the effective recovery of 46% compared to conventional mechanical

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stirring. Öncel et al. [4] have reported silver recovery of 96.6% from solid residue using ultrasound-assisted thiourea leaching process.

Thiourea (TU) as an reagent for extracting silver has shown broad prospects for implementation in the metallurgical industry [18,19]. Laboratory experiment has indicated that thiourea leaching process for silver extraction has several advantages including lower toxicity, environment friendly, easier handling of reagent, greater selectivity for silver, and fast leaching kinetics over the cyanidation leaching process [20].

In this study, the XRD and SEM are utilized to identify the difference in morphology and phase constitution comparing ultrasonic-assisted leached residue and conventional leached residue. The effects of the particle size, thiourea concentration, reaction temperature, on the leaching rate have been evaluated. The leaching kinetics of silver in aqueous sulfuric acid solution was examined according to the shrinking core model. Additionally the order of the reaction and the activation energy were estimated.

2. Materials and methods

2.1. Materials

The SD used in this study was collected from Kunming Iron & Steel Holding Co., Ltd. (KISC). The chemical compositions of washed SD is listed in Table 1. The crystalline phases of the samples were investigated. Fig. 1a shows presence of hematite (Fe_2O_3), magnetite (Fe_3O_4), cotunnite ($PbCl_2$), laurionite (PbOHCl), anglesite ($PbSO_4$), quartz (SiO_2), and calcia (CaO) as the main crystals in the mineral phases of the sample. SEM image of the washed SD was observed by a scanning electron microscope, shown in Fig. 1b.

2.2. Experimental procedure

Prior to use, the sintering dust was washed with tap water at room temperature for 60 min at a pulp density of 200 g/L with mechanical stirrer at 400 r/min to remove the soluble substances. After filtration, the residue was dried and sieved. The leaching experiment with ultrasonic augmentation and conventional leaching were carried out under atmospheric pressure, in a beaker of 500 ml volume which was placed on a thermostatically controlled water bath equipped with magnetic stirring, having precision of ±1 °C. Dilute sulfuric acid was utilized as solvent with thiourea to vary the concentrations of acidic thiourea solution.

40 g of sample was taken in a beaker with pH = 1.0–1.5 acidic thiourea solution at a ratio of liquid to solid 8:1. The reaction temperature was preset by thermostatically controlled water bath. Compared with the conventional leaching process, the mechanical stirrer is replaced with an ultrasonic transducer in ultrasound-augmented leaching process. The air was fed into the reaction vessel for a duration of 5 min, intermittently at an interval of 10 min. The ultrasonic waves were generated by ultrasonic transducer (Nanjing Han Zhou Technology Co., Ltd, in China) with a frequency of 20 kHz at varying power ranging from 200 to 1000 W. The schematic diagram of the conventional and the ultrasound-augment leaching experimental setup is shown in Fig. 2.

The dependent variables are varied in the following range: the ultrasound power from 20% to 80% of the total power 1000 W, the particle size from 75 μm to 830 μm, and the leaching temperature in the range of 25–50 °C, the thiourea solution concentration between 10 and 22 g/L. After the process of leaching, the filtrate and residue were analyzed. The yield (Y, %) of silver was calculated according to following formula:

$$Y = \frac{X_0 - X_i}{X_0} \times 100\% \quad (1)$$

where X_0 represented the silver content in water washed SD, X_i was the silver content of the residue.

2.3. Analytical methods

The mineralogical composition and crystallinity of the washed SD was performed on powdered samples using a Rigaku D/Max 2500/PC powder diffractometer. Chemical analyses were performed on whole powders by X-ray fluorescence (Bruker AXS S4 Pioneer). The micro-morphological characteristics of washed SD

Table 1
Principal elements in the washed SD (XRF data).

Element	Fe_2O_3	PbO	CaO	SiO_2	SO_3	Cl	F	Al_2O_3	MgO	Bi_2O_3	Ag[a]
Content (wt.%)	52.99	19.09	6.41	5.34	5.08	1.99	1.40	1.28	1.26	0.96	699 g/t

[a] The content of Ag was analyzed using ICP-AES.

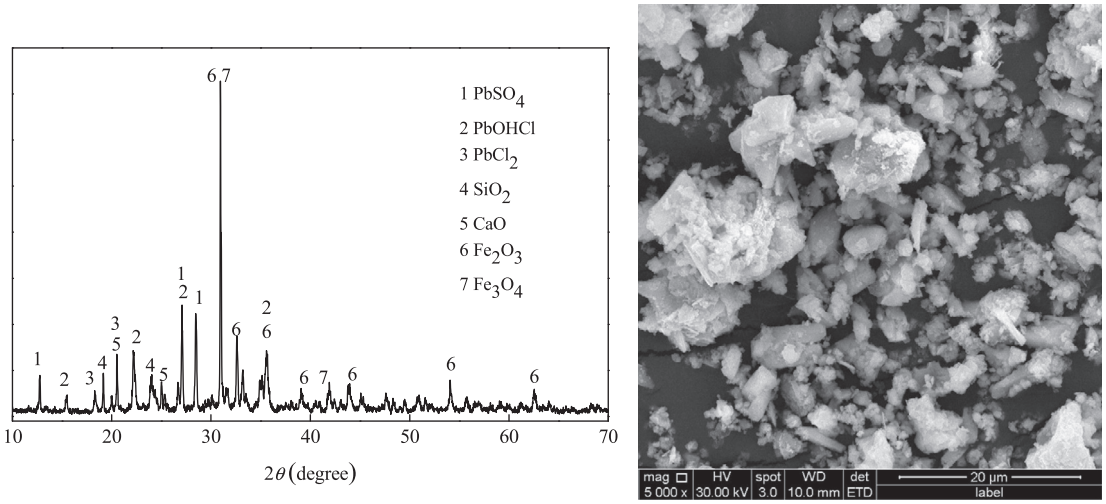

Fig. 1. XRD pattern and SEM image of the washed residue of SD.