Separation and Purification Technology 115 (2013) 66–72

Contents lists available at SciVerse ScienceDirect

Separation and Purification Technology

journal homepage: www.elsevier.com/locate/seppur

Leaching kinetics of zinc residues augmented with ultrasound

Xin Wang [a,b,c], C. Srinivasakannan [d], Xin-hui Duan [f], Jin-hui Peng [a,b,c,*], Da-jin Yang [e], shao-hua Ju [a,b,c]

[a] Kunming University of Science and Technology, Faculty of Metallurgy and Energy Engineering, Kunming, Yunnan 650093, China
[b] Key Laboratory of Unconventional Metallurgy, Ministry of Education. (Kunming University of Science and Technology), Kunming, Yunnan 650093, China
[c] Engineering Laboratory of Microwave Application and Equipment Technology, Kunming, Yunnan 650093, China
[d] Chemical Engineering Program, The Petroleum Institute, P.O. Box 2533, Abu Dhabi, United Arab Emirates
[e] Kunming Metallurgy Research Institute, Kunming, Yunnan 650093, China
[f] Institute of Power Source & Eco-materials Science, Hebei University of Technology, Tianjin 300130, China

ARTICLE INFO

Article history:
Received 28 November 2012
Received in revised form 15 April 2013
Accepted 23 April 2013
Available online 30 April 2013

Keywords:
Zinc
Zinc plant residues
Leaching kinetic
Ultrasound-assisted
Shrinking core model

ABSTRACT

The leaching kinetics of zinc residue, having total Zn content of 12.31%, along with other metallic components such as Fe and Pb, is leached using sulfuric acid as solvent, augmented with ultrasound is presented. The effects of variables such as the leaching temperature, sulfuric acid concentration, particle size, liquid/solid ratio and the ultrasound power have been assessed. The results show the maximum recovery of zinc to be 80% at an ultrasound power of 160 W, leaching temperature of 65 °C, sulfuric acid concentration of 1.4 mol/L, particle size range of 74–89 μm and liquid/solid ratio of 4. The kinetics of leaching is modeled using shrinking core model and the rate controlling step is identified to be the diffusion through the product layer. The raw and the leached residue are characterized using XRD and SEM/EDX analysis. The activation energy is estimated to be 6.57 KJ/mol, while the order of reaction with respect to sulfuric acid concentration is 0.94 and particle size is 0.12 respectively.

1. Introduction

Zinc is one of the most popular and versatile metal that finds wide application, including plating, coating and alloying with other metals. Commercially zinc can be rated as the third most common nonferrous metal after aluminum and copper [1–3]. Currently, the vast majority of metallic zinc is produced by the Roast–Leach–Electro-winning (RLE) process [4]. This process generates large amount of leach residue as insoluble substance, which still contains significant amount of zinc [5]. The zinc residue is categorized as hazardous waste, further creating disposal problems to the operating plants [6].

The high demand for zinc has attracted the interest of industry to utilize the secondary sources such as zinc ash, zinc dross and leach residues as potential valuable sources [7]. In the process of leaching, many insoluble materials are concentrated in the residue, while the zinc is present in the form of zinc ferrite [8]. Altundoğan et al. and Safarzadeh et al. [9] have reported recovery of the valuable metals from the zinc plant residue. Additionally, the kinetics of zinc recovery from the residue using sulfuric acid dissolution has also been reported in the literature [10–13]. It has been reported that the high temperature and high acid concentration can increase the leaching rate, and the activation energy for these reactions are reported [14–16]. However, ultrasound assisted zinc leaching has been reported rarely in open literature.

Although RLE process is industrially established, the accumulation of insoluble substances on the ore surface restrains the liberation of zinc ions. Over the past decade, different unconventional ways of zinc recovery have been attempted, which include application of microwave [17], ultrasound [18], pressure [19], mechanical [20] methods. The use of ultrasound is reported to increase the leaching yield as well as shorten the duration of leaching due to the well-known cavitation effect, rendering it to be a popular method in the metal processing industry. Avvaru et al. [21] have reported a significant increase in the leaching efficiency with ultrasound as compared to conventional mixing methods in the process of extraction of uranium (an radioactivity element) from Narwapahar uranium ore with nitric acid and sulfuric acid being the leaching agents. Similarly, Hursit et al. [22] have reported enhanced zinc leaching kinetics from smithsonite ore with ultrasound. Swamy and Narayana [23] have designed dual frequency ultrasonic leaching equipment which was reported to increase the recovery by 46% as compared to a conventional agitation process. Öncel et al. [24] have reported a 96.6% yield of silver from solid residue using microwave assisted leaching with thiourea. A 20% increase in the % yield of TiO_2 was reported from red mud as compared to conventional leaching methods under identical conditions by Sayan and Bayramoğlu [25]. The following compilation well evidences the effectiveness of ultrasound assisted leaching as compared to

* Corresponding authors at: Kunming University of Science and Technology, Faculty of Metallurgy and Energy Engineering, Kunming, Yunnan 650093, China. Tel.: +86 57 18687117030.

E-mail address: wangxinwa84@163.com (W. Xin).

1383-5866/$ - see front matter
http://dx.doi.org/10.1016/j.seppur.2013.04.043

conventional process. Bese [26] have reported % yields for ultrasound-assisted leaching to be 89.28% for copper, 51.32% for zinc, 69.87% for cobalt, and 1.11% for iron vs. 80.41% for copper, 48.28% for zinc, 64.52% for cobalt, and 12.16% for iron, in the absence of ultrasound. An overall increased effectiveness of ultrasound assisted leaching can be inferred in comparison with the conventional leaching process.

In the present study, the effects of parameters such as ultrasound power, leaching temperature, particle size, sulfuric acid concentration and liquid/solid ratio were assessed on the leaching kinetics of zinc. The kinetic data was modeled using the popular Shrinking Core Model (SCM), which accounts for chemical reaction and product layer diffusion control. In addition the order of the reaction was estimated, using a semi-empirical equation, for the diffusion-controlled process. The leached samples were characterized using the advanced analytical instruments such as X-ray diffraction (XRD) and scanning electron microscope (SEM–EDS) [27].

2. Materials and methods

2.1. Materials

The zinc residues used for the experiments were supplied by a metallurgical plant of Yunnan province, China. Prior to the experiments, the samples were washed, dried and sieved to different size ranges. X-ray diffraction analysis (XRD) of the zinc residue shows the mineralogical content of the sample (Fig. 1).

XRD analysis shows the $ZnFe_2O_4$ and $PbSO_4$ and SiO_2 as the major components in the sample. The chemical composition of different size ranges of zinc residues were also analyzed using an atomic absorption spectrometer and the results are shown in Table 1 [28].

2.2. Methods

The ultrasound-assisted leaching experiments were carried out in a 1000 mL, lidded beaker in a thermostatically controlled water bath, within a precision of ±0.1 °C, equipped with sampling device. The ultrasonic transducer was positioned inside the beaker and connected to an ultrasonic generator which generates ultrasonic waves with a frequency of 20 kHz at different power outputs of 80, 160, and 240 W (Guangzhou Hengda Ultrasonic Electric Technological Ltd., China). A schematic of the experimental setup is shown in Fig. 2.

Table 1
Chemical analysis of different sieve fractions of zinc residues.

Serial number	Size (μm)	Content (%)	Element (%)			
			Zn	Fe	Pb	SiO_2
1	104–89	9	12.68	21.22	12.26	6.99
2	89–74	11	12.80	20.36	12.38	6.95
3	74–61	16	12.48	21.75	12.50	7.03
4	61–53	18	12.36	21.64	12.19	7.00
5	<53	30	12.58	21.15	12.07	7.10

Deionized water was utilized as a diluent with sulfuric acid to prepare various concentrations of acid solution. The dependent variables were varied in the range: acid solution/zinc residues weight ratio (3–6), the acid concentration (0.3–1.7 mol/L), the ultrasound power (80–240 W), the particle size range (<53 to 89–104) and the leaching temperature (55–85 °C). The rate of leaching was estimated by withdrawing a small sample (<5 mL). The zinc content of filtered sample was estimated using atomic absorption spectrometer, and the experimental variations were less than ±2%. The Zn recovery was estimated, using the equation:

$$Zn_{recovery} = \frac{Zn_{T_0} - Zn_T}{Zn_{T_0}}$$

The analysis of solid residue was carried out using SEM–EDS (XL30ESEM-TMP scanning electron microscope, Philips, Holland) and XRD (Brukerd-advance diffractmeter, Germany) with a step size of 0.02°, ranging from 10° to 90°at 30 mA and 40 kV.

3. Results and discussion

Reaction between zinc ferrite and sulfuric acid can be written as follows [11,29–32]:

$$2ZnFe_2O_4 + 8H_2SO_4 \rightarrow 2Fe_2(SO_4)_3 + 2ZnSO_4 + 8H_2O \quad (1)$$

Based on the above reaction, the effects of variables ultrasound power, leaching temperature, acid concentration, particle size and liquid/solid ratio on the leaching rate were assessed.

3.1. Kinetics analysis

Solid–fluid heterogeneous reactions are common in chemical and hydrometallurgical processes. In order to determine the kinetic parameters and rate controlling step of Zn leaching process, the popular shrinking core model was utilized [33].

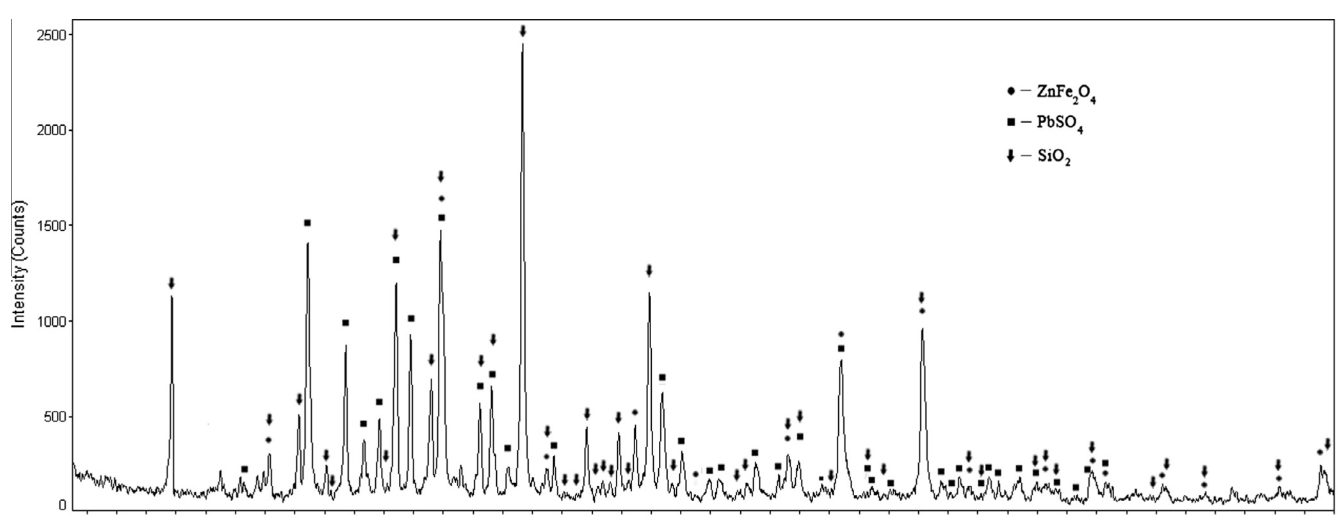

Fig. 1. XRD patterns of zinc plants residues.