Chemical Engineering and Processing 97 (2015) 75–83

Contents lists available at ScienceDirect

Chemical Engineering and Processing: Process Intensification

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

Kinetics of microwave roasting of zinc slag oxidation dust with concentrated sulfuric acid and water leaching

Jun Chang[a,b,c], Li-bo Zhang[a,b,c], Chang-jiang Yang[a,c], Qianxu Ye[a,b,c], Jing Chen[d,*], Jin-hui Peng[a,b,c,**], Chandrasekar Srinivasakannan[e], Wei Li[b]

[a] State Key Laboratory of Complex Nonferrous Metal Resources Clean Utilization, Kunming University of Science and Technology, Kunming 650093, China
[b] Key Laboratory of Intensification Metallurgy in Yunnan Province, Kunming University of Science and Technology, Kunming 650093, China
[c] Faculty of Metallurgical and Energy Engineering, Kunming University of Science and Technology, Kunming 650093, China
[d] School of Chemical Science and Technology, Yunnan University, Kunming 650091, China
[e] Chemical Engineering Program, The Petroleum Institute, P.O. Box 2533, Abu Dhabi, United Arab Emirates

ARTICLE INFO

Article history:
Received 24 March 2015
Received in revised form 28 July 2015
Accepted 3 September 2015
Available online 7 September 2015

Keywords:
Microwave roasting
Zinc slag oxidation dust
Indium
Kinetics

ABSTRACT

The microwave irradiation is used to roast zinc slag oxidation dust in the presence of concentrated sulfuric acid with the aim of accelerating roasting process. The roasted material is leached with tap water for the recovery of zinc and indium. The influence of roasting temperature, particle size, and the ratio of acid to ZSOD mass on the rate of roasting reactions were investigated. The results show that the extraction rate of zinc and indium increases with increasing the roasting temperature and the ratio of acid to ZSOD mass, while decreases with increase in the particle size. The samples were characterized using XRD and SEM. Under the conditions employed in the present work a maximum recovery of 94% and 90% of zinc and indium could be achieved. Shrinking core model was used to describe the kinetic parameters and to identify the rate controlling step. Both zinc and indium extraction were controlled by the internal diffusion in the solid product layer, with the apparent activation energy of the roasting reaction were 15.94 kJ/mol and 12.68 kJ/mol respectively. The reaction order with respect to concentrated sulphuric acid was 1.75 and 1.59 for zinc and indium, respectively.

1. Introduction

Zinc leach residue, a solid waste from zinc hydrometallurgical process, usually contains significant proportion of low grade zinc and indium. Hence it is pertinent to ensure recovery these metals in view of economic and environmental consideration [1–3]. The conventional acid leaching process would consume large amount of solvents, additionally generating large volumes of wastewater streams, rendering the recovery process not feasible [4]. Rotary kiln reduction is popularly adopted in many zinc plants to enrich indium and zinc from zinc leach residue through oxidation of zinc slag dust [5]. The zinc slag oxidation dust(ZSOD) also contains some stable compounds like zinc ferrite which wrap certain proportion of metal indium [6–9], rendering unsatisfactory recovery. Recent efforts to develop cleaner and efficient processes to recover zinc and indium from the ZSOD include different ways, such as hot sulfuric acid leaching [9–12], Na_2CO_3 roasting [13], the pressure oxidative acid leaching [14], high-energy planetary mill to modify the chemical stability of indium-bearing zinc ferrite [15].

Microwave heating has many characteristics, such as selective, uniform and fast heating, no pollution, low equipmentcost, very fast reaction speed and high product yields [16]. Microwave roasting and extraction of minerals in metallurgy has been a subject of intense interest in these years [17–18]. It is being increasingly applied to improve the yield of extracted metals and to reduce processing duration, especially with demand for more environmental benign processes. It has been well documented that recovery of metals from industrial residues can be increased significantly with application of microwave energy compared to conventional heating [19–22], which can be attributed to the mode of heating (uniform) and enhanced rates of heating [23].

Application of microwave roasting extraction is reported to refractory gold ore [17], vanadium extraction from stone coal [24], carbothermal reduction of zinc ferrite [25], zinc recovery from EAF dust mixed with carbon [26]. However application of microwave roasting for ZSOD, is at its primitive with very limited reports in

* Corresponding author.
** Corresponding author at: State Key Laboratory of Complex Nonferrous Metal Resources Clean Utilization, Kunming University of Science and Technology, Kunming 650093, China.
E-mail address: JingChen_ynu@163.com (J. Chen).

http://dx.doi.org/10.1016/j.cep.2015.09.006
0255-2701/

open literature. The present work proposes a novel process to improve the recovery of zinc and indium from ZSOD through application of microwave roasting.

The possible chemical reactions taking place during sulphation roasting of ZSOD can be written as follows [27]:

$$2ZnFe_{(2-m)}\quad In_mO_4+16H_2SO_4=2ZnSO_4+(4-2_m)\quad Fe_2(SO_4)+2_mIn_2(SO_4)+8H_2O \tag{1}$$

$$ZnO+H_2SO_4=ZnSO_4+H_2O \tag{2}$$

$$PbO+H_2SO_4=PbSO_4+H_2O \tag{3}$$

$$CaO+H_2SO_4=CaSO_4+H_2O \tag{4}$$

The process conditions such as roasting temperature, particle size and the ratio of acid to ZSOD mass were assessed in the present work. Furthermore, the microwave roasting kinetics of zinc and indium leaching from ZSOD with concentrated sulfuric acid provide a theoretical guide for the process optimization and industrial application in the future.

2. Materials and methods

2.1. Materials

The ZSOD received from a zinc metallurgical plant in Yunnan province, China. Prior to experimentation the samples were dried and sieved to different size ranges. X-ray diffraction analysis (XRD) of the ZSOD (Fig. 1a) show that the main mineralogical phases includes zincite (ZnO), lead oxide (PbO), franklinite ($ZnFe_2O_4$), magnesium silicate (Mg_2SiO_4), and so on. Different morphology and aggregation can be observed in Fig. 1b. The main elements is presented in Table 1, which was tested by Kunming Metallurgical Research Institute, China.

2.2. Experimental procedure

The roasting was carried out in self-made microwave oven which had a temperature control system along with a vacuum pump to remove the gases formed during roasting. Firstly, necessary water was put into 100 g ZSOD to wetting the sample enough in a clean ceramic crucible. Then, a certain amount of concentrated sulphuric acid was added to the sample and stirred uniformly. At last, the ceramic crucible was placed in the microwave oven for roasting. The experiments were repeated covering the various process conditions. The design of experiment was represented in Table 2. After being roasted the sinter was weighed, ground, and immersed in a sealed glass bottle for leaching with water at room temperature. The solid–liquid mixture was magnetically stirred at a rotational speed of 300–350 rpm for 30 min. After solid/liquid separation and the washing of leach residue, the concentration of indium and zinc in leached residue was characterized by atomic absorption spectrometry (Z2000HITACHI). The% leaching (x) of zinc or indium was calculated according to the following equation:

$$x=\frac{m_1\times c_1-m_2\times c_2}{m_1\times c_1}\times 100\%=\left(1-\frac{m_2\times c_2}{m_1\times c_1}\right)\times 100\% \tag{5}$$

where m_1 and m_2 is the mass of ZSOD and leached residue, and c_1 and c_2 is the concentration of zinc or indium containing in ZSOD and leached residue, respectively.

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

Components	Zn	Pb	Fe	S	Ca	As	Sn	In
% wt	34.22	17.70	11.78	2.88	2.03	1.51	1.40	0.15

Table 2
The design of experiment

Parameters	Levels						
Time (min)	0	10	20	30	40	50	60
Temperature (°C)	90		120		150		180
Average particle size (μm)	53		66.5		90.5		128
Mass ratio of acid to ZSOD	0.2:1		0.3:1		0.4:1		0.5:1

2.3. Characterization

The phase transitions of the sample were identified using XRD technology (D/Max 2200, Rigaku, Japan). XRD patterns were recorded using Rigaku diffractometer with Cu Ka radiation and a Ni filter operated at the voltage of 35 kV, anode current of 20 mA and a scanning rate of 0.25° min^{-1}, respectively. The microstructure morphology of the samples was investigated by scanning electron microscopy (SEM). The SEM instrument (XL30ESEM-TMP, Philips, and Holland) was operated at 20 kV in a low vacuum.

3. Results and discussion

3.1. Effect of temperature

The effect of roasting temperature on % extraction of zinc and indium from ZSOD with concentrated sulfuric acid is assessed at

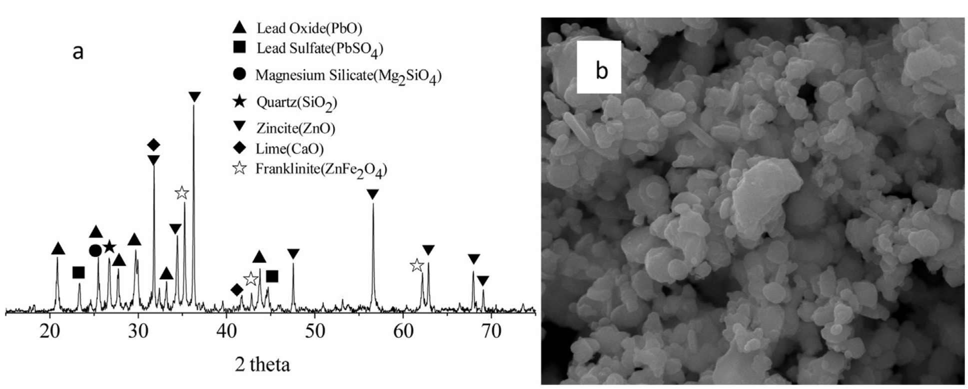

Fig. 1. XRD pattern (a) and SEM image (b) of zinc slag oxidation dust.