



Pretreatment of tin anode slime using alkaline pressure oxidative leaching



Junwei Han, Chao Liang, Wei Liu^{*}, Wenqing Qin^{*}, Fen Jiao, Wenhua Li

School of Minerals Processing and Bioengineering, Central South University, Changsha 410083, China

ARTICLE INFO

Article history:

Received 11 July 2016

Received in revised form 25 October 2016

Accepted 25 October 2016

Available online 1 November 2016

Keywords:

Alkaline pressure oxidative leaching

Arsenic removal

Crystallization

Evaporation

Tin anode slime

ABSTRACT

The pretreatment of tin anode slime using alkaline pressure oxidative leaching was studied. The effects of different parameters on the extractions of various metals were investigated based on thermodynamic calculation. The results indicated that the optimum leaching conditions were 2.5 M of NaOH concentration, 130 °C of temperature, 1.5 h of time, 1 MPa of oxygen partial pressure, 7 mL/g of liquid-to-solid ratio and 700 rpm of stirring speed. Under the conditions, the extractions of tin and arsenic reached 92.16% and 96.49% respectively, but that of copper and lead were at a low level. Meanwhile, antimony, bismuth, and silver were hardly dissolved into the NaOH solution. The leachate was then subjected to an evaporation and cooling crystallization process for arsenic removal. After the purification, 94.36% of arsenic was separate from the lixivium as sodium arsenate hydrate and the final solution can be used to produce sodium stannate.

© 2016 Elsevier B.V. All rights reserved.

1. Introduction

Tin anode slime, as a byproduct, is produced from electrolytic refining process of crude tin and contains many valuable metals, such as Sn, Pb, Sb, As, Cu, Bi, Ag, Au, and In, which are in the form of insoluble phases and finally precipitate in the bottom of refining tanks. It is economically attractive and is regarded as one of the important materials for the extraction of precious metals [1]. However, the arsenic impurity markedly reduces its economic value due to the requirements of additional treatments for avoiding the containment of hazardous emissions during the recycle of metals by pyrometallurgical process.

In general, the process for the treatment of anode slime includes the following steps: pretreatment, concentration, and refining, among which pretreatment is the most crucial to determine the whole process. A number of methods have been studied for the pretreatment of tin anode slime, which can be summarized as follows: oxidizing roasting-acid leaching [2–4], beneficiation-metallurgy combination process [5–8], reduction smelting-electrolysis [9], vacuum carbothermal reduction [10]. However, these methods present many problems, such as serious secondary arsenic pollution, extensive energy consumption, and high difficulty in separation of tin and antimony. Therefore, it is necessary

to explore an economical and eco-friendly method to pretreat tin anode slime, especially for the material containing high content of arsenic and antimony.

Compared to these methods, pressure leaching is cleaner and more efficient, and has been extensively applied in the processing of nickel [11], zinc [12], cobalt sulfides [13,14], and refractory gold concentrate [15,16], etc. But there are few relevant reports on the pretreatment of tin anode slime with pressure technology in alkaline medium. However, the pretreatment of copper and lead anode slime with pressure leaching have been studied in recent years, which can be used for reference. Liu et al. [17] used the process of alkaline pressure oxidative leaching (APOL) to selective separate arsenic and selenium from copper anode slime with the extractions of arsenic and selenium reaching up to 99.0% and that of lead, copper, tellurium and silver tending to 0%. Yang et al. [18] also obtained good results by the same technology to pretreat lead anode slime and the leaching rates of arsenic and lead were 98% and 4.0%, respectively.

In the present study, an APOL was developed to selectively extract tin and arsenic from tin anode slime, and the removal of arsenic from the leach solution was carried out by an evaporation and cooling crystallization process. The effects of NaOH concentration, temperature, oxygen partial pressure, leaching time and liquid-to-solid (L/S) ratio on the extractions of tin and arsenic were investigated based on thermodynamic calculation. The final solution and solid products were analyzed by ICP and XRD for confirming the experimental results.

^{*} Corresponding authors.

E-mail addresses: ase.6520@163.com (W. Liu), qinwenqing369@126.com (W. Qin).

2. Experimental

2.1. Materials

The tin anode slime used in this study was supplied from a tin smelter in China. The original tin anode slime was firstly dried at 100 °C and then sieved with 0.4 mm sieve to remove large particles. The main chemical composition of the tin anode slime is given in Table 1. It can be seen from Table 1 that the tin anode slime contains high antimony and arsenic, which is essential to remove arsenic in advance for preventing arsenic secondary pollution. The X-ray diffraction (XRD) pattern of the sample shown in Fig. 1 indicates that the antimony contained in the anode slime is mainly in the form of metallic antimony (Sb) and mopungite [NaSb(OH)₆]. There was no other mineral phase identified from the anode slime due to their amorphous nature. In addition, sodium hydroxide (NaOH) of analytical grade and oxygen gas (O₂) with 99.9% purity were used in the pressure leaching experiments.

2.2. Experimental setup and procedure

A schematic diagram of the experimental setup is presented in Fig. 2. The leaching experiments were conducted in a 2 L stainless steel autoclave, which was equipped with an oxygen tank for providing continuous and stable oxygen partial pressure. Temperature and stirring speed were controlled by a proportional integral derivative controller. For each test, 100 g of anode slime powder and 700 mL of NaOH solution with a scheduled concentration were firstly mixed and poured into the autoclave. Then the autoclave was sealed and heated at a rate of 5 °C/min. When heating to the required temperature, open the valve of the oxygen tank to control the pressure inside the autoclave. After a given time, turn off the controller of temperature and the inlet valve, and open the cooling water valve for cooling the autoclave to about 70 °C. Thereafter, open the outlet valve and the cover of autoclave. At the end of each experiment, the slurry in the autoclave was pumped and vacuum-filtrated, and then washed three times with tap water. The leaching residue was dried, weighed, and grinded, followed by digestion and chemical composition analysis. The filtrate was measured for volume using a measuring cylinder and sampled for chemical composition by ICP, and was subjected to an evaporation and cooling crystallization process in a 250 mL glass beaker for the removal of arsenic by filtration. The crystals produced were then dried at 80 °C to prevent loss of crystal water, and the metal compositions of the solution and crystals were analyzed by chemical method and/or ICP. Additionally, an electric hot plate was used in the evaporation process.

2.3. Analytical methods

The contents of metals were detected with different methods according to their amounts in solution or residue. Sb, Bi, Pb, Cu, Au, Ag, In, Si and the low contents of Sn and As were detected by inductively coupled plasma-atomic emission spectrometry (ICP-AES, IRIS Intrepid II XSP), after the sample powders were digested with a mixture of concentrated HCl and HNO₃ (3:1, v/v) [19–21]. The high contents of Sn and As were analyzed by chemical method with iodine and potassium bromate. In addition, the crystalline

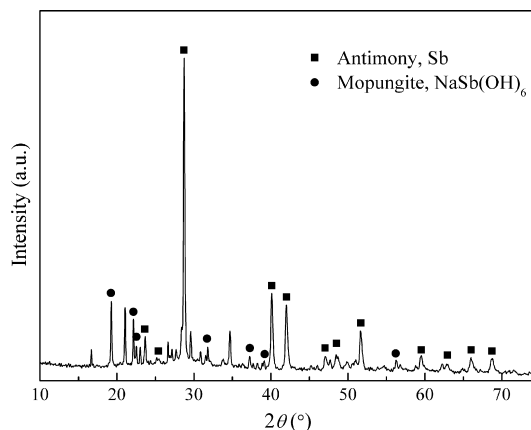


Fig. 1. XRD pattern of the tin anode slime.

compositions of the samples were analyzed using X-ray powder diffraction (XRD, Germany Bruker-axs D8 Advance) [22].

3. Results and discussion

3.1. E-pH diagrams

The Metal-H₂O systems were used as a guide in pH and potential values of the leaching situation. Fig. 3 gives the E-pH diagrams of As-H₂O, Sn-H₂O, Pb-H₂O and Cu-H₂O system at 150 °C. Note that the concentrations of related metallic ions are fixed at 1 M, and both of the partial pressures of oxygen and hydrogen are at the standard atmospheric pressure of 101,325 Pa. The E-pH diagram of As-H₂O system shown in Fig. 3(a) indicates that an increase in potential will lead to the conversion of As → As(III) → As(V). The reaction of As(V) in aqueous solution can be regarded as a complex formation between AsO₄³⁻ and hydrogen ion. In alkaline solution, the concentration of hydrogen ion is so low that most of As(V) exists as monomeric AsO₄³⁻. In Sn-H₂O system, the transformation of Sn → Sn(IV) occurs in the whole pH range investigated. In alkaline solution, Sn(IV) exists as SnO₃²⁻, and it tends to precipitate as Sn(OH)₄ and further dissolve as Sn⁴⁺ with the decrease in pH, as shown in Fig. 3(b). It can be seen from Fig. 3(c) and (d) that lead and copper can be oxidized to soluble species in acid solution and to insoluble species in neutral and alkaline solution. In strong alkaline solution, excessive hydroxyl ions can dissolve copper, lead and their oxides. In addition, other valuable metals, such as Sb, Ag, Au, Bi and In, are probably exist as insoluble species in alkaline leaching but dissolve in acid solution, based on our knowledge. Therefore, it is found that alkaline leaching is an effective method to selectively extract Sn and As from tin anode slime.

3.2. Alkaline pressure oxidative leaching

3.2.1. Effect of NaOH concentration

The chemical composition of the raw material shows that it contains about 47.94% Sn and 10.25% As. The selective extraction of Sn and As with NaOH solution under pressure was studied. The possible chemical reactions during the leaching process are listed as follows:

Table 1
Main chemical composition of the tin anode slime.

Composition	Sn	As	Sb	Bi	Pb	Cu	Au	Ag	In	Si
Wt.%	47.94	10.25	16.73	4.8	5.92	3.9	0.002	0.15	0.34	0.43

Download English Version:

<https://daneshyari.com/en/article/4990200>

Download Persian Version:

<https://daneshyari.com/article/4990200>

[Daneshyari.com](https://daneshyari.com)