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A novel method to recover zinc and iron from zinc leaching residue

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ABSTRACT

A novel method to recover zinc and iron from zinc leaching residue (ZLR) by the combination of reduction roasting, acid leaching and magnetic separation was proposed. Zinc ferrite in the ZLR was selectively transformed to ZnO and Fe_3O_4 under CO, CO_2 and Ar atmosphere. Subsequently, acid leaching was carried out to dissolve zinc from reduced ZLR while iron was left in the residue and recovered by magnetic separation. The mineralogical changes of ZLR during the processes were characterized by XRF, TG, XRD, SEM–EDS and VSM. The effects of roasting and leaching conditions were investigated with the optimum conditions obtained as follows: roasted at 750 °C for 90 min with 8% CO and CO/CO + CO_2 ratio at 30%; leached at 35 °C for 60 min with 90 g/l sulfuric acid and liquid to solid ratio at 10:1. The iron was recovered by magnetic separation with magnetic intensity at 1160 G for 20 min. Under the optimum operation, 61.38% of zinc was recovered and 80.9% of iron recovery was achieved. This novel method not only realized the simultaneous recovery of zinc and iron but also solved the environmental problem caused by the storage of massive ZLR.

1. Introduction

Currently, more than 85% of the world's metal zinc is produced by the conventional hydrometallurgical approach including oxidative roasting, acid leaching, purification, and electrowinning processes (RLPE) (Jha et al., 2001; Turan et al., 2004). During the oxidative roasting process, ZnS is converted to ZnO, but a significant fraction reacts with the iron impurities to form zinc ferrite (Dimitrova et al., 2000; Langová et al., 2009; Peng et al., 2012; Vahidi et al., 2009). Zinc ferrite is hardly soluble in mild acidic conditions, and a huge quantity of leaching residue is produced in the subsequent leaching process. The residues could be used to recover metallic values or it may be disposed off (Soner Altundoğan et al., 1998).

Pyrometallurgical and hydrometallurgical processes have been studied to recycle zinc and iron from ZLR. The most typical pyrometallurgical method is Waelze process, and the recovery of zinc could reach to 90–95% in this process. However, Waelze process consumes huge energy to maintain the high reaction temperature (above 1300 K), and large amount of high iron-bearing secondary residue is generated (Beşe et al., 2010). These shortcomings also exist in other pyrometallurgical methods including Ausmelt technology (Hoang et al., 2009), roasting with Na_2CO_3 (Holloway et al., 2007), roasting with Na_2S (Zhang et al., 2011), and some other methods (Antrekowitsch, 2001; Beşe et al., 2010; Çoruh and Nuri, 2010). The most common hydrometallurgical process is to recycle zinc and iron from ZLR in a hot concentrated sulfuric acid followed by precipitation of the dissolved iron values from solution as jarosite, goethite or hematite (Graydon, 1988). High leaching rate both of zinc and iron could be obtained in these processes, but the removal of iron and other unwanted metals from the leaching solution is difficult (Dimitrios Filippou, 1997). Hydrochloric acid leaching (Langová et al., 2009), alkaline solution leaching (Dutra et al., 2006; Zhao and Stanforth, 2000), microwave caustic leaching (Xia and Picklesi, 2000), D2EHPA leaching (Vahidi et al., 2009), and leaching with various solvents have been studied (Langová et al., 2007). These hydrometallurgical processes are more economical because of lower capital and operating costs, but the purification process becomes more complex. Therefore finding a cost-effective and environmental-friendly process to recover zinc and iron from zinc leaching residue remains the major challenge.

In this study, a simultaneously recycling zinc and iron from ZLR by the combination of selective reduction, acid leaching and magnetic separation is investigated. The objective of this recycling technology is to decompose $ZnFe_2O_4$ to ZnO and Fe_3O_4 selectively. Then, ZnO and Fe_3O_4 could be separated by acid leaching and magnetic separation, respectively. This study focus on the various operating parameters in the three stages under laboratory conditions to control phase transformation and search the optimum experiments conditions. The phase transformation, morphology variation and magnetism changes of the samples are detected by

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XRD, SEM and VSM to interpret some phenomenon observed in experiments and the decomposition procedure of $ZnFe_2O_4$.

2. Experimental approach

2.1. Materials and analysis

ZLR used in this study was obtained from a zinc hydrometallurgical plant in Hunan province, China. The samples were dried at 105 °C for 24 h before grinding and sieved to obtain required fractions. Elements composition of the residue was determined by XRF and results in Table 1 show that the main contents in ZLR used in this study are Zn-19.57%, Fe-23.91%, S-6.40%, Si-4.47%, Pb-4.35%, Ca-2.21%. The crystalline phases of the samples were investigated by X-ray powder diffraction (Rigaku, TTR-III) in 2θ scale using Cu Kα radiation (λ = 1.5406 Å, 50 kV and 100 mA) at the scanning rate of 10°/min vary from 10° to 80°. As Fig. 1 shows that zinc ferrite ($ZnFe_2O_4$), magnetite (Fe_3O_4), zinc sulfide (ZnS), zinc silicate (Zn_2SiO_4), and calcium sulfate ($CaSO_4$) are the main crystal mineral phases in the residue. The morphological changes were detected by Scanning Electron Microscopy (SEM, JEOL.LTD, JSM-6360LV). Vibration sample magnetometer (VSM) (Model: HH-15) was applied to assess the magnetism of magnetic concentrates. Potassium dichromate titration and EDTA titration methods were used to quantitative analysis the proportion of ferrous in total iron and content of zinc oxide to evaluate the effect of experiments (Zhang, 1992).

2.2. Experimental procedure

2.2.1. Reduction roasting

The experiments were performed according to the flow sheet as shown in Fig. 2. Reduction roasting process was firstly operated in thermo-gravimetric apparatus (STA449F3) to decompose zinc ferrite to ZnO and Fe_3O_4 selectively. The ZLR was flatted on the inner wall of the silica crucible and pick up the suction. Then, the sample was heat up to determined temperature at 10 K/min under 100 ml/min Ar. As soon as the temperature reached, a proportion fraction of reduction gases (CO + CO_2 + Ar) was added to take place of Ar at 100 ml/min, respectively. After reduction roasting, the mixed reduction gases (CO + CO_2 + Ar) were changed to Ar again to cool down the samples. The reduced products were stored in a tightly closed jar waiting for analysis.

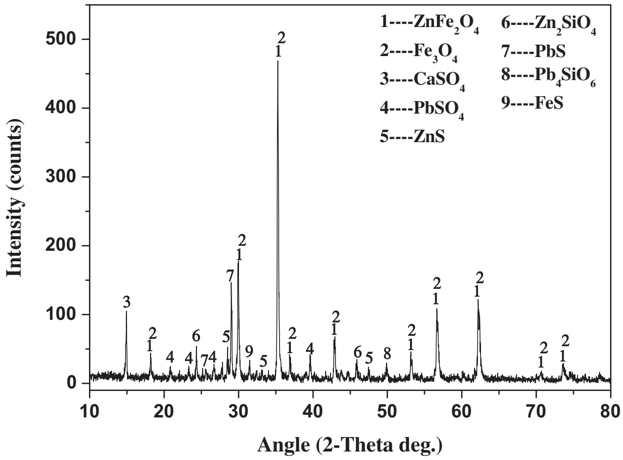

Fig. 1. X-ray diffraction pattern of ZLR.

Table 1
Chemical composition of ZLR by XRF.

Elements	Content (wt.%)	Elements	Content (wt.%)	Element	Content (wt.%)
Fe	23.91	Cu	0.8	Cl	0.067
Zn	19.57	As	0.52	Ti	0.057
S	6.40	Cd	0.31	In	0.054
Si	4.47	Mg	0.26	P	0.043
Pb	4.35	K	0.24	Ag	0.037
Ca	2.21	Ba	0.20	Cr	0.018
Mn	1.53	Sr	0.16	Ni	0.011
Al	1.15	Sn	0.16	Mo	0.010

The influences of reduction roasting parameters such as CO concentration, CO/CO + CO_2 ratio, roasting temperature and duration time were studied as detailed shown in Table 2. The content of zinc

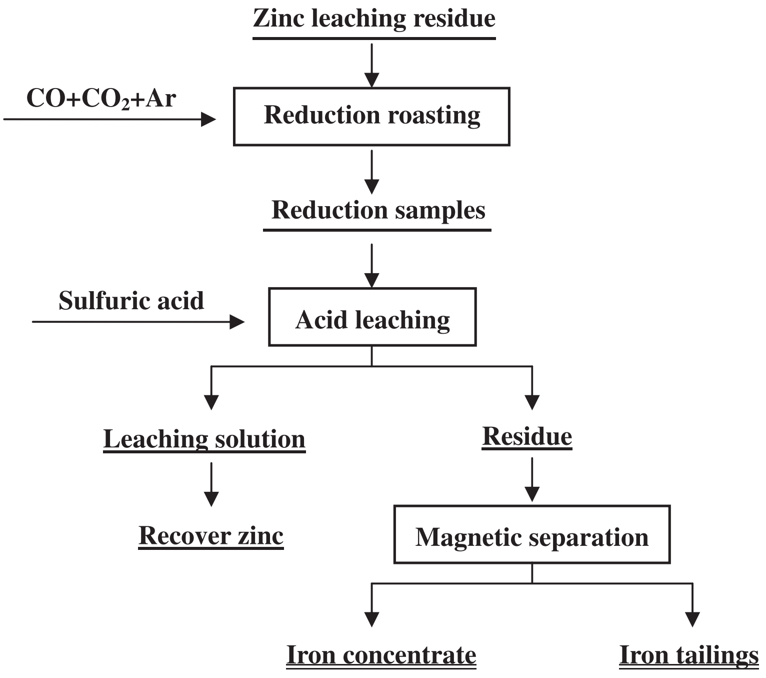

Fig. 2. Flow sheet of the process of simultaneously recovering zinc and iron from ZLR.