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Silver leaching and recovery of valuable metals from magnetic tailings using chloride leaching



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ABSTRACT

To comprehensively reuse lead-zinc tailings, the recovery of valuable metals from magnetic tailings after iron recovery from lead-zinc tailings using roasting followed by chloride leaching was studied in this paper. The recovery of metals was investigated by studying the effects of roasting temperature, leaching temperature, liquid-to-solid ratio, chloride ion concentration and acid concentration on the metal leaching efficiency. The results showed that the leaching efficiencies of Ag, Ga and Pb reached 84.39%, 80.76% and 70.47%, respectively, at a roasting temperature of 900 °C and with the optimized leaching parameters. The Ag (9.98 mg/L), Ga (18.62 mg/L) and Pb (1506.12 mg/L) in the leaching liquor could be further recovered. The leaching kinetics showed that the silver leaching was controlled by the diffusion of the fluid species, and the activation energy was 22.06 kJ/mol. Furthermore, the leaching residue was analyzed. The results of X-ray fluorescence spectrometry, chemical speciation and extraction toxicity analyses suggested that the pollution risk of the leaching residue has been substantially eliminated and that further reuse of the residue was possible. With this method, the processes of metal recovery from magnetic tailings using roasting followed by chloride leaching were proposed for the comprehensive utilization of lead-zinc tailings.

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1. Introduction

The increasing amount of lead-zinc tailings in China poses a serious risk to the environment and human health. To comprehensively use these lead-zinc tailings, the sulfur and iron are recovered from the lead-zinc tailings, and magnetic tailings are produced (Lei et al., 2017, 2015). As a result, many metals, including silver and other valuable metals, are enriched in the magnetic tailings during the magnetic separation. Due to the non-degradation and persistence of metals in the environment, the metals in magnetic tailings are highly toxic and carcinogenic when they are transferred to stream water, groundwater and soil (Akcil and Koldas, 2006). The recovery of metals from tailings is one of the most effective methods to avoid pollution by metals while achieving an economic benefit (Chen et al., 2014). Additionally, enrichment of the valuable metals in the magnetic tailings makes the tailings a significant resource (Lei et al., 2017).

As an efficient technology of hydrometallurgy, leaching is the most used method to recover silver and other valuable metals. To recover silver from tailings, various reagent systems, including chloride (Zhang et al., 2016), cyanide (Hernandez et al., 2014), thiourea (Li et al., 2012) and thiosulfate (Amaral et al., 2014) media, have been tested. However, cyanide leaching is limited because of increasing concerns regarding the toxicity of cyanide (Johnson, 2015). Thiourea is also a suspected carcinogen (Örgül and Atalay, 2002). In addition, the high price and instability of thiourea and thiosulfate hinder their wide application in silver recovery. Compared with these techniques, chloride leaching is attractive in terms of low cost, mild toxicity and technical maturity (Hasab et al., 2014). Moreover, in chloride leaching, the formation of chloride complexes with different valuable metals makes it easier to achieve a comprehensive recovery of metals in tailings.

Many experimental investigations have been carried out on silver recovery from silver-bearing wastes through chloride leaching (Yazici and Deveci, 2015; Zhang et al., 2012). However, the recovery of silver from low-grade tailings, especially from magnetic tailings, is rarely reported. Moreover, some tailings need oxidative





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pre-treatments (roasting, bacterial leaching or pressure leaching) in order to improve recoveries and achieve economic feasibility (Puente-Siller et al., 2013). However, research about the effects of oxidation on the chloride leaching of silver and other metals is lacking. In addition, the transformation of other metals during the leaching process is an important factor that not only affects the further recovery of silver from the leach liquor but also determines the potential environmental pollution of the leaching residue. Due to the complexity of the metals in magnetic tailings, investigations of silver leaching and the metal transformation during the oxidation roasting and leaching processes are important and necessary. More importantly, leaching residue is a key concern because of the possible risk of environmental pollution. However, the current studies on leaching pay little attention to environmental risk assessment of the leaching residue.

To avoid environmental pollution and recover silver and other valuable metals from magnetic tailings, the oxidation roasting and chloride leaching of magnetic tailings were studied in this paper. The effects of roasting temperature, leaching temperature, liquidto-solid ratio, chloride ion concentration and acid concentration on the silver recovery were investigated, and the kinetics of silver leaching were analyzed. The leaching characteristics of other metals were discussed as well. Furthermore, analysis of the leaching residue was performed to investigate potential environmental pollution and the possibility of further reuse. It is expected that this paper will provide an effective method for recovering metals in magnetic tailings from lead-zinc tailings and solve the environmental pollution problem of lead-zinc tailings.

2. Materials and methods

2.1. Materials and experimental procedures

The raw material used in this study was the magnetic tailings obtained from a previous study (Lei et al., 2017, 2015), in which magnetic tailings were produced from a series of treatment processes (roasting and magnetic separation) of the lead-zinc tailings from the Fankou lead and zinc mine, Renhua, Guangdong, China. The samples were ground to below 200 mesh and dried at 105 ± 2 °C for 24 h.

Oxidation roasting of the sample was carried out in a muffle furnace. Samples were placed inside the furnace at a specified temperature for 30 min, taken out and then cooled. Their weights were measured by an electronic balance (Sartorius BSA124S-CW, Germany). The leaching experiment was carried out isothermally at different temperatures and for various durations in separate batch experiments using a glass reactor with a magnetic stirrer (DF101S, Jintan Medical Instrument Plant, China). All the chemical reagents used in this study were analytically pure. The leaching efficiency of the metal was calculated as follows:

$$E = \frac{c_l V}{c_s m} \times 100\% \tag{1}$$

where *E* is the leaching efficiency of the metal; c_s is the metal content of the solid sample treated by leaching; *m* is the mass of the solid sample; c_l is the metal concentration of the leaching liquor; and *V* is the volume of the leaching liquor.

2.2. Analytical methods

The mineral phases of zinc were determined using a chemical analysis method (Nonferrous Metals Industry Analysis Series Editorial Board, 1992). According to this method for determining zinc mineral phases, a sequence of extractants (H₂O, ammonium

acetate, acetic acid and saturated bromine water) was used to extract the different mineral phases of zinc (ZnSO₄, ZnO, ZnSiO₃ and ZnS). The residue was analyzed to determine the ZnFe₂O₄ content. The chemical speciation of the metals in the tailings and the leaching residue were analyzed based on the modified BCR threestep sequential extraction procedure (Nemati et al., 2009). According to the procedure, metals were defined in the following four chemical fractions: acid soluble fraction (F1), reducible fraction (F2), oxidizable fraction (F3) and residual fraction (F4). The extraction toxicity of the leaching residue was determined using a solid waste-extraction procedure for leaching toxicity - sulfuric acid & nitric acid method (SEPA, 2007a). According to the method, the process that the extracts harmful components from wastes into the environment under the influence of acid precipitation was simulated using a mixture of nitric acid and sulfuric acid as the extraction agent. The concentrations of heavy metals in the extraction solution should meet the requirement of national standard.

To determine the metals in the solid samples, the samples were digested using microwave digestion equipment (WX-8000; Shanghai Yi-Yao Instruments, Shanghai, China) according to method 3052 (US EPA, 1996). The metals in solution, including Fe, Pb, Zn, Mn, Ag and Ga, were determined by an atomic absorption spectrophotometer (AAS, Hitachi ZA3000). The mineralogical phase analysis was carried out using an X-ray diffractometer (XRD, MiniFlex 600, Rigaku). An X-ray fluorescence spectrometry (XRF, Rigaku 100e) was used for multi-element analysis. To ensure the accuracy of the analysis results, all the above tests were performed in triplicate, and the average of these test results was used as the final value.

3. Results and discussion

3.1. Characteristic of the tailings

The XRD pattern of the magnetic tailings is depicted in Fig. 1. As shown in Fig. 1, the major components in the magnetic tailings are SiO₂ and Fe₃O₄. The Fe₃O₄ was the portion that was not extracted during the magnetic separation and remained in the magnetic tailings. The chemical composition of the magnetic tailings was determined as listed in Table 1. The content of silver in the tailings was 56.33 g/t. In addition, the other metals in the magnetic tailings included Ga, Pb, Zn and Mn. It is apparent that it could be profitable to recover these metals from magnetic tailings. The main impurity elements included calcium, aluminum and silicon.

Chloride leaching was used to recover silver from the tailings. However, for the magnetic tailings, an oxidative pre-treatment was necessary in order to improve the leaching of silver (Rodríguez-



Fig. 1. XRD pattern of the tailings.

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