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Production of lead concentrate from bioleached residue tailings by brine leaching followed by sulfide precipitation

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ABSTRACT

This study investigated a process for the production of lead (Pb) concentrates from bioleached residue by brine leaching, followed by sulfide precipitation of lead sulfides. Bioleached residue characterization indicated that the Pb was in the form of anglesite ($PbSO_4$). The bioleached residue was extracted by leaching it in a sodium chloride (NaCl) solution at room temperature for 200 r/min. This effectively extracted Pb. More than 84% of the lead was extracted from the bioleaching residues at a NaCl concentration of 150 g/dm^3. Pb concentrate was quantitatively produced from the brine leaching solution by adding sodium sulfide as a precipitant. The optimum conditions for Pb precipitation were: 0.6 g/dm^3 sodium sulfide (Na_2S); and 10 min leaching time in the solution. X-ray diffractometer (XRD) or scanning electron microscopy (SEM) analysis was used characterize the raw materials, brine leached residues, and precipitation. The study also investigated the mechanism involved in brine leaching and precipitation of Pb from the bioleached residue. Finally, a scale-up experiment found that total Pb extraction exceeded 88%, and the quality of the Pb concentrate could meet industrial requirements for roasting processes. This process provides an eco-friendly and economic alternative for treating solid waste containing $PbSO_4$.

1. Introduction

As the primary source of Pb, high grade sulfide ores are steadily becoming depleted. Recovering Pb from mine tailing or low-grade ore is recognized as a needed alternative to meet future demand. However, it is expensive to use traditional processes to recover Pb from mine tailings or low-grade ore.

Different methods are used to recover metals from mine tailing, including acid leaching and bioleaching. Each method has its own limitations. Compared with physical and chemical processes [1,2], bioleaching is relatively inexpensive, safe, simple to operate, and environmentally friendly [3,4]. Many studies have been conducted to apply bioleaching techniques to extract metals from Pb ores, slag, and concentrates [5–7]. During the bioleaching process, solubilized Pb^{2+} has formed $PbSO_4$ ($K_{sp} = 1.8 \times 10^{-8}$) with sulfate; this material has low solubility, making lead extraction inefficient. However, many bioleached residues are generated in the bioleaching process. Poorly soluble lead (II) sulfate ($PbSO_4$) forms during the bioleaching stage, and leads to a high concentration of Pb in the bioleached residue. As such, a process to improve the leaching rate of Pb after bioleaching is needed. Furthermore, approaches to recover metals from bioleached residues have been developed in mines to reduce environmental problems as well as creating a new source [8].

Several recovery methods have been applied to study different industrial wastes containing Pb. These include alkaline leaching [9,10], acid leaching [11,12], and brine leaching [8,13]. However, brine leaching (using NaCl, $MgCl_2$, and $CaCl_2$) is the most recognized and widely used method to recover Pb [8]. Compared with Pb other recovery methods, brine leaching is relatively simple to operate and environmentally friendly.

Abdollahi et al. [14], Turan et al. [15] and Farahmand et al. [8] applied brine leaching to enrich lead ($PbSO_4$) in zinc plant residues. Liao and Deng [16] extracted lead using combined sequential bio-oxidation and acidic brine leaching from raw complex sulfide ores. Guo et al. [17] used brine leaching to recover metals from hydrometallurgical residue. More than 90% of the lead was subsequently extracted from the residue.

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Chloride-based leaching systems have advantages over sulfate-based systems. Most metal chlorides are considerably more soluble than sulfate salts. It is easy to regenerate brine leaching reagents using cyclic processes [8]. Lead sulfate is insoluble ($K_{sp} = 1.8 \times 10^{-8}$) in water, but is soluble in saturated chloride solutions. Brine leaching is based on the formation of complex chlorides of lead [8]. Brine leaching can promote the leaching efficiency of heavy metals, especially in recovering lead [15]. Studies focusing on brine leaching to extract Pb from bioleached tailings residue are significant, because they offer a new method to recover Pb.

After the leaching process, the metal is transferred from the solid waste to the leachate, and remains in the leaching solution. This highlights the need to separate/remove the valuable metal from the leaching solution. Many previous studies have focused on removing metal from electronic waste, leaching residue, mine ore, ash, slag, or tailing, but have not addressed recovering metals from leachate [11,13,18–20]. Metals are removed from the leachate using extraction, electrowinning, or precipitation [9,21,22]. After recovering the valuable metals, the solutions are recycled for leaching.

Hydroxide precipitation is widely used in industry to remove metals [23,24]; sulfide precipitation is not used as widely. This may be because sulfide dosing is considered difficult to control. Sulfide precipitation has some advantages, including lowering the solubility of metal sulfide precipitates, increasing the potential for selective metal removal, increasing reaction rates, improving settling properties, and increasing the potential to re-use sulfide precipitates through smelting [24]. Metal sulfide precipitation is a significant process for hydrometallurgically treating ores and effluents [24]. Past studies to explore this achieved a total extraction of Pb, Cu, Ni, and Zn of 80% or more [9,25,26]. Further, the quality of metal concentrates can meet industrial requirements for roasting processes [9]. Metal sulfide precipitation can effectively recover heavy metals in the leaching solution.

This study investigated an integrated process for producing Pb concentrates from bioleached residue tailing by brine leaching, followed by sulfide precipitation. The study's first stage investigated lead extraction; the bioleached residual material from the lead-zinc mine tailing was leached using a sodium chloride (NaCl) solution. The second stage explored a process to precipitate lead concentrates from the brine leaching solution using sodium sulfide (Na_2S).

2. Material and methods

2.1. Bioleached residue tailings sampling and analysis

Bioleached residue tailing was collected from lead-zinc mine tailings after bioleaching. More than 97% of the zinc had been extracted by the bioleaching process; residue samples were isolated using a 0.45 μm filter membrane and a vacuum pump. The bioleached residue was washed with a sulfuric acid solution (pH 2.0) and distilled water. The residue was then crushed and dried in a vacuum drying box. The samples were then used for the NaCl leaching experiments. Bioleached residue particles used for brine leaching tests were less than 74 μm in size.

The samples were digested using microwave digestion equipment (CEM Mars-xpress) to determine metal concentrations using a flame atomic absorption spectrophotometer (FAAS; Hitachi Z-2000). The morphology and structure of the bioleached residue before and after brine leaching were examined using scanning electron microscopy (SEM; Hitachi S-3400N(II)) and an X-ray diffractometer (XRD; Rigaku Ultima-IV) analysis. The bioleached residue contained approximately 0.723% Pb. Fig. 1 shows the XRD patterns and SEM images of the bioleached residue.

2.2. Brine leaching experiments

After the bioleaching process, the bioleached residue was subjected to brine leaching to assess Pb recovery. Leaching of the bioleached residue was conducted in a beaker placed on a stirrer-unit. The experiment also investigated the interactions between pulp density (10–300 g/dm^3) and sodium chloride concentration (10–150 g/dm^3). The effects of leaching time (20 s–30 min) were examined at room temperature and 200 r/min, using a 150 cm^3 beaker containing a 100 cm^3 clear brine solution. After each run, the brine leaching slurry was immediately filtered through a 0.45 μm membrane. The leaching solution sample was analyzed for Pb.

2.3. Leaching solution precipitation experiments

These specific experiments investigated the impact of Na_2S dosage and reaction time on Pb precipitation. Each experiment was conducted at room temperature, with a leached solution volume of 50 cm^3. The Na_2S doses added to the brine leaching solution ranged from 0.2 to 0.7 g/dm^3. Each sample was continuously agitated at a speed of 200 r/min for 20 min. The effect of reaction time on Pb precipitation was examined by setting the reaction time at 1, 2, 5, 10, 15, 20, and 30 min, with a fixed Na_2S concentration of 0.6 g/dm^3, and a fixed agitation speed of 200 r/min. The residual total Pb concentration in the solution was measured using a FAAS after the solution was filtered through a 0.45 μm membrane.

2.4. Chemical analysis

During the leaching experiment, the Pb concentrations in the leaching solutions were measured using FAAS. XRD was used to characterize the mineral phase composition in the bioleached residue, brine leached residue, and precipitates. After the samples were freeze-dried and coated with gold, SEM was used to evaluate the mineral surface morphologies. The Pb concentrate was measured using Methods of Chemical Analysis of Lead Concentrates [27].

The percent removal of Pb in brine leaching process was calculated using Eq. (1):

$$\text{Pb recovery } (\%) = M_2/M_1 \times 100\% \quad (1)$$

In the expressions, M_1 is the mass of Pb solubilized in brine leaching solution (mg); M_2 is the mass of Pb in the bioleached residue (mg).

The Pb precipitation efficiency was calculated using Eq. (2):

$$\text{Pb precipitation } (\%) = (C_1C_2)/C_1 \times 100\% \quad (2)$$

In this expression, C_1 is Pb concentration in the leaching solution (g/dm^3); and C_2 is Pb concentration in the supernatant after precipitation experiments (g/dm^3).

3. Results and discussion

3.1. Brine leaching for Pb recovery

Pb recovery efficiency was low after the bioleaching [28]. Lead sulfate is insoluble in water, but becomes soluble in saturated chloride solutions. As such, the study investigated the interactions between NaCl concentration and pulp density, and the effects of leaching time on Pb recovery from bioleached residue.

3.1.1. Effects of pulp density and NaCl concentration on Pb recovery

As Fig. 2 shows, Pb recovery increased significantly when the NaCl concentration increased. When the NaCl concentration