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Removal of metals from lead-zinc mine tailings using bioleaching and followed by sulfide precipitation

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HIGHLIGHTS

- Bioleaching combined sulfide precipitation is a feasible method to remove metals.
- FTIR and XRD confirmed the composition of tailing was changed during bioleaching.
- Sulfide precipitation can remove metals in bioleaching solution.
- Kinetic is controlled by shrinking core model after some time had elapsed.

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ABSTRACT

Mine tailings often contain significant amounts of metals and sulfide, many traditional operations used to minerals was not as good as those currently available. This study investigated metals removal from lead-zinc mine tailings using bioleaching and followed by sulfide precipitation. Metals were dissolved from the tailings by the bacteria in a bioleaching reactor. During a 10% pulp density bioleaching experiment, approximately 0.82% Pb, 97.38% Zn, and 71.37% Fe were extracted after 50 days. With the pulp density of 10% and 20%, the dissolution of metals followed shrinking core kinetic model. Metals (Pb, Zn, and Fe) present in the pregnant bioleaching leachate. Metals were next precipitated as a sulfide phase using sodium sulfide (Na_2S). Metal precipitations were selectively and quantitatively produced from the bioleaching leachate by adding Na_2S. More than 99% of the zinc and 75% of the iron was precipitated using 25 g/L Na_2S in the bioleaching leachate. The results in the study were to provide useful information for recovering or removing metals from lead-zinc mine tailings.

1. Introduction

Mining and metallurgical activities generates considerably amounts of mine tailings. Most tailings have been left in tailing ponds or mine sites without treatment. These have a long-term potential threat to engineering and environmental (Falagán et al., 2017). Mine tailings contain many sulfide minerals, and sulfide minerals are a combination of sulfide with metals (iron, copper, nickel, lead and zinc). These can produce acid mine drainage (AMD), and cause damage to environmental (Falagán et al., 2017; Lei et al., 2017). Heavy metal mobilizations and acid generation into the surrounding environment lead to soil and groundwater contamination and decreased biological diversity (Liu et al., 2008). These outcomes endanger both the ecosystem and human health. Therefore, to avoid those problems, suitable, effective, and economic technologies are needed to recover or remove heavy metals from mine tailings.

To recover or remove metals form mine tailings, systematic research was investigated by different processes including roasting, magnetic separation, and leaching (Chen et al., 2014a; Lei et al.,

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2017; Liu et al., 2008). Bioleaching technology is widely used to recover or remove metals from mine ores (Olson et al., 2003; Pradhan et al., 2008). Bioleaching is relatively low cost, high safety, simple to operate, and environmentally friendly (Chen and Lin, 2009; Pradhan et al., 2008). Bioleaching has been increasingly used to extract and recover heavy metals from solid waste. Target materials include concentrates, ore, mine tailings, electronic waste, and sediments (Zhu et al., 2011; Seidel et al., 2006; Ghassa et al., 2014; Cheng et al., 2009; Mohanty et al., 2017). Bioleaching is based on the ability of acidophilic chemolithotrophic microorganisms, using elemental sulfur or Fe^{2+} as energy sources, to dissolve sulfide minerals in the form of extractable metals, these metals can then be recovered or removed.

Most previous studies focused on removing the metal from the mine ores, slag, or tailing, but without recovering metals from leachate (Hong et al., 2014; Li et al., 2014; Marrero et al., 2015; Falagán et al., 2017). Few studies have explored the recovery of metals from the bioleaching leachate (Liao and Deng, 2004). This highlights the need to separate/remove other metal ions in order to recover valuable metals from leachate. The metals are removed from the leachate using solvent extraction, electrowinning, cementation, or precipitation (Mokone et al., 2012; Chen et al., 2016; Mishra et al., 2016). After recovering the valuable metals, the solutions are recycled for leaching.

The recovery of lead from lead-zinc tailings was examined in the previous study, and the results indicated that the technology for the recovery of lead from lead-zinc tailings was feasible using the bioleaching combination of bioleaching with brine leaching and followed by sulfide precipitation (Ye et al., 2017a,b). However, leachate also contained many hazardous heavy metals. As the waste product from bioleaching, the leachate leads to an environmental problem. It need a useful method to remove metal from leachate.

Metal sulfide precipitation is a significant process for hydrometallurgically treating ores and effluents. Hydroxide precipitation is widely used in industry to remove metals (Theiss et al., 2016). However, sulfide precipitation is not used as widely as hydroxide precipitation, perhaps because sulfide dosing is considered difficult to control. Previous studies have shown that 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 (Lewis, 2010). A series of sulfide precipitation studies (Lewis and van Hille, 2006; Liu et al., 2011; Mokone et al., 2010) was conducted and revealed that the total extraction of lead, copper, nickel, and zinc reached or exceeded 80%. Further, metal concentrate qualities can meet industrial requirements for roasting processes. Metal sulfide precipitation can effectively recover the heavy metals in the leaching solution.

This study investigated the removal of metals from mine tailings using bioleaching and sulfide precipitation combination. To this end, the first stage investigated metals extraction using a bioleaching reactor. The second stage of the study explored the feasibility of a process to precipitate heavy metal from the bioleaching leachate. The study investigated the effects of the added amount of sodium sulfide on the precipitation of metal. The goal of the study was to provide useful information for recovering or removing metals from lead-zinc mine tailings.

2. Methods

2.1. Mine tailings sample

Lead-zinc tailings used for this study were collected from a lead-zinc mine plant in Shaoguan, China. Lead-zinc mine tailings were prepared by crushing and screening the material to particle sizes below 74 μm. A representative sample of the mine tailings was pulverized to conduct the chemical and mineralogical analysis. The pH of the tailing was determined using a solution of tailings and water at a 1:2.5 ratio (tailings/water (w/v)) (Marrero et al., 2015). The solid samples were digested using microwave digestion equipment (CEM Mars-xpress) to determine metal concentrations. Table 1 provides the chemical analysis of the tailings.

2.2. Bacteria culture and bioleaching experiments

To support this study, a local iron-oxidizing bacterium (*Acidithiobacillus ferrooxidans*) was isolated and purified from an acid mine drainage in a lead-zinc mine region. The bacteria were grown and maintained in the 9 K medium. The cultures were grown in a shaken flask culture (30 °C and initial pH 2.0) before being used as the inoculum in bioleaching experiments. The experiments were carried out in 3 L reactor, containing 2 L iron-free 9 K medium at an inoculation ratio of 10 (i.e., ratio of bacterial solution to culture medium of 1:10). The initial bacterial concentration in the bioleaching medium was determined in a cell counting chamber using phase-contrast microscopy (Olympus CX41), and kept constant at 1×10^7 cells/mL.

Prior to the bioleaching experiment, bioleaching in initial pH of 2.0 had a good leaching efficiency to recover metals. The reactor was agitated at room temperature, an initial pH of 2.0, and a pulp density of 10% or 20%. The reactors were stirred continuously at 200 rpm with an electric stirrer. During the experiments, aliquots were periodically collected from each flask to determine pH and heavy metals concentrations. Water lost during the bioleaching process through evaporation was replenished with distilled water daily. The pH of the medium was adjusted to 2.0 with H_2SO_4.

2.3. Sulfide precipitation experiments

These specific experiments investigated the impact of dosage of sodium sulfide on metals precipitations. Each experiment was conducted at room temperature, with a leachate volume of 50 mL. The dosage of Na_2S added in bioleaching leachate was 2.5–30 g/L, and then continuously agitated at a speed of 200 r/min in the stirrer for 20 min. The residual total metal concentration in the solution was determined using an atomic absorption spectrophotometer (AAS; Hitachi Z-2000) after filtering through a 0.45 μm filter.

2.4. Chemical analysis

During the leaching experiment, Cu, Fe, Pb, and Zn concentrations in the solutions were determined using AAS. The pH values and redox potential Eh were measured using a pH meter and redox potential analyzer (Ohaus Starter 2100). Mine tailings were mixed in KBr by triturating in a mass ratio of 1:99 and then were pressed

Table 1
Chemical composition of the mine tailings.

Parameter	Concentration
Cu(mg/kg)	220
Fe (mg/kg)	128 405
Pb (mg/kg)	6293
Zn (mg/kg)	5653
S_{total} (mg/kg)	128 940
pH	7.80
Organic matter (%)	3.22
Water content (%)	0.80