

An Experimental Study of Acid Leaching Efficiency for Metal Recovery from Copper Tailing

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Abstract

The effect of the different H₂SO₄ addition was investigated under a fixed S: L of 1:2. The leaching temperature was set to 40°C, and leaching occurred for 2 h. The leaching efficiency of the metals increased gradually with an increasing amount of H₂SO₄. Through AAS method, the leaching efficiency of 0.99±0.09% and 1.00±0.03% for Cu was obtained with the addition of 1.5 and 2 mL/g of H₂SO₄. For Fe, leaching efficiencies of 13.95±1.42% and 11.70±1.22% were obtained with the addition of 1.5 and 2 mL/g of H₂SO₄. However, 17.57±2.81% and 15.63±2.86% for Mn was leached when 1.5 and 2 mL/g of H₂SO₄ was added. Statistically, there was non-significant ($p<0.05$) difference in leaching efficiency of Cu, Fe and Mn among the different concentrations of H₂SO₄. Using XRF method, the leaching efficiency of 0.03±0.01% and 0.04±0.01% for Cu was obtained with the addition of 1.5 and 2 mL/g of H₂SO₄. For Fe, leaching efficiencies of 11.13±2.89% and 12.80±3.38% were obtained with the addition of 1.5 and 2 mL/g of H₂SO₄. However, 0.18±0.10% and 0.19±0.09% for Mn was leached when 1.5 and 2 mL/g of H₂SO₄ was added. Statistically, there was non-significant ($p<0.05$) difference in leaching efficiency of Cu, Fe and Mn among the different concentrations of H₂SO₄. It is concluded that Cu, Fe and Mn from the low grade copper tailing are successfully recycled via a H₂SO₄ leaching and subsequent fractional precipitation technology. Based on the mineral composition analysis results, leaching temperature, the amount of H₂SO₄ added, leaching time, and S:L have been evaluated for the leaching efficiencies. The amount of H₂SO₄ added and S:L have strongly influenced the leaching efficiency.

Keywords

Copper Tailing; Acid Leaching; Heavy Metals.

1. Introduction

A common environmental issue associated with the mineral industries is the disposal of a huge mass of tailing materials regularly produced from their processing operations. Historically, mining industries do not have a good reputation because of releasing their waste materials to the surrounding environment. Tailings dams as a practical solution have played an important role in protecting valuable soil and water resources from contaminated slurries. Tailings dams are considered the largest man-made structures in the world. They are generally comprised of three types of materials: (1) factory sediments (2) mine tailings (3) deposited materials. Normally, considerable bodies of water may be stored behind tailings dams, so dam failure can cause disastrous damages to lives, properties, and the surrounding environment.

Copper tailings (Cu tailings) present a serious environmental problem in many regions worldwide. When exposed to natural weathering and chemical percolation, heavy metals, including Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn, become more soluble and mobile. Once leached, heavy metals would become the major sources of contamination, which seriously damage not only stream water and groundwater, but also the soil environment. Meanwhile, a large amount of tailings presents a significant metal resource. In order to solve these problems, metal recovery through the separation method has been studied, the separation depends upon those independent mineral features such as magnetism gravity and palatability [1]. However, it is impossible to achieve a comprehensive recovery [2]. On the other hand, with leaching methodology, metals such as Cu, Zn, Fe, Ni, and Mg could be recycled and the recovery efficiencies were much higher than that for the separation process [3]. Although methods of hydrometallurgical leaching for sulfide ores as well as for some wastes, such as batteries have been reported [4]. Few studies have been reported of its application for the Cu tailing. Recycled metals using the leaching process have to be purified for further reuse [5].

Mining is the most obtrusive of man's modification of the environment and mining activities are well known for their deleterious effect on the environment. A harmful effect of mining activities is reflected in the degradation of large areas of land (open pits), disposal of large volumes of solid waste and the occurrence of acid mine drainage [6]. This is typical of copper extraction from sulfide ores for instance, in the Attic Copper Mine in northern Sweden, which is one of the biggest copper mines in Europe, the mass of the extracted copper per ton of ground ore is only 3 kg, whereas the average Cu content in the ore is 0.4%, which is about the world average [7]. In copper recovery from such low-grade sulfide ores, it is customary to use hydrometallurgical processes (leaching, solvent extraction, and electrowinning) or beneficiation is performed by froth flotation (including crushing, grinding, flotation, thickening, and drying of the concentrate), which is followed by pyro metallurgical treatment of the concentrate and electrolysis [8].

It has been reported that the precipitation was an effective method for it is operationally simple and economical. With the precipitation process, the metals were enriched as a mixture in the sludge, while the metal content of the sludge was too low for reuse [9]. However, the metal content of the sludge would be promoted by the fractional precipitation process. Thus, effective leaching with a fractional precipitation method for the recovery of valuable metals from Cu tailing is still highly desired. As a typical poly-metallic mine in Southern China, the Dabaoshan Mine region is one of the largest opencast copper mining bases [10]. The amount of Cu tailing generated is approximately 480,000 tons per year, and the amount of tailings stored in the tailing reservoir is approximately 800 million tons. The reservoir becomes a source of acid mine drainage (AMD) contaminating the well water around the Dabaoshan Mine region. Therefore, comprehensive reuse of the tailing would improve economic efficiency and lessen environmental damage as well. Such comprehensive reuse could be promoted by the leaching process. The objective of this work is to recover Cu, Fe, Zn, and Mn from low-grade Cu tailing effectively using an environment-friendly and economical leaching method [11].

Copper tailing is the solid waste material left during the purification of the precious copper from the copper ores. Disposal of copper tailing is one of the major important environmental issues in a copper mine. Mining of metals generates considerably amounts of waste materials. These generally have very little economic value, making their exploitation not profitable, though they often have the potential to pose a long-term threat and cause damage to the environment.

2. Experimental measurements

This present research was laid out to study the metal recovery from copper tailing through acid leaching. For this purpose, the solid material samples were brought into the Environmental resources laboratory. The solid material samples were crushed into the pestle and mortar. H₂SO₄, HCl and H₂O were added to prepare the samples. The samples were placed into shaker in a way to get them homogeneous for about different times. The homogenized samples were filtrated with the help of a Water-Circulation multifunction vacuum pump machine in a way to get liquid filtrate samples. After

the liquid filtrate was tested by AAS analysis and reduced solid residue samples were tested under XRF for their elemental analysis. The expected outcome result was show us the percentage of leached material and the determination of heavy metal and their total contents were studied. At first, the pH of the leaching filtrate was adjusted with the addition of $\text{Ca}(\text{OH})_2$ to recover Fe. Thereafter, Cu was recovered through addition of Na_2S solution. Finally, Mn was recovered by adjusting the pH in the leaching filtrate.

The solid material samples were brought into the environmental resources laboratory. The solid material samples were crushed into the pestle and mortar. H_2SO_4 , HCL and H_2O were added to prepare the samples. The samples were placed into shaker in a way to get them homogeneous for about different times. The homogenize samples were filtrated with the help of a Water-Circulation multifunction vacuum pump machine in a way to get liquid filtrate samples. After the liquid filtrate was tested by AAS analysis and reduced solid residue samples were tested under SEM & EDS for their elemental analysis. The expected outcome result was show us the percentage of leached material and the determination of heavy metal and their total contents were studied. At first, the pH of the leaching filtrate was adjusted with the addition of $\text{Ca}(\text{OH})_2$ to recover Fe. Thereafter, Cu was recovered through addition of Na_2S solution. Finally, Mn was recovered by adjusting the pH in the leaching filtrate.

The solid samples were digested by microwave digestion equipment (WX-8000, EU Microwave Chemistry Technology Co., Ltd.) according to method 3052. In this section, the major metals were determined using an AAS (Shimadzu AA6300, made in Japan), whereas minor metals at significantly low concentrations were determine through ICP-MS (Agilent 7500, made I America). The mineral composition was investigated by SEM-EDX and analyzed through the SIP system of QEMSCAN. The bound moisture content was determined through the method of specific gravity.

The parameters affecting the recovery efficiency, such as leaching temperature, amount of added leaching reagent, leaching time, and leaching solid-to-liquid ratio (S:L, weight to volume) was investigated. The fractional precipitation was used for the metal separation recovery from the leachate, whose principle is the solubility discrepancy of different metals. The metal recovery from the leaching filtrate was preceded by fractional precipitation with the addition of different chemical agents.

The statistical analysis was performed by SPSS and Origin software. The three replicates of each sample were analyzed for their statistical analysis. The results were expressed as the arithmetic means \pm standard deviation. Statistical data was analyzed using Microsoft Excel 2007 and SPSS 19.0. One-way analysis of variance (ANOVA) was used to detect the statistical significance of differences between groups of data with a significance level of $p < 0.05$.

3. Results and discussion

Toxic mining waste is currently a problem of higher concern both for industries and the government. There are a number of copper tailings dumps that have been abandoned and not sufficiently stabilized. Once the tailings containing toxic minerals and residual metals come in contact with diluents like rain water, it forms acid mine water that can leach metals into ground water, rivers and streams.

The effect of the different H_2SO_4 addition was investigated under a fixed S: L of 1:2. The leaching temperature was set to 40°C , and leaching occurred for 2 h. As presented in Table-1 and 2, the leaching efficiency of the metals increased gradually with an increasing amount of H_2SO_4 . Through AAS method, the leaching efficiency of $0.99 \pm 0.09\%$ and $1.00 \pm 0.03\%$ for Cu was obtained with the addition of 1.5 and 2 mL/g of H_2SO_4 . For Fe, leaching efficiencies of $13.95 \pm 1.42\%$ and $11.70 \pm 1.22\%$ were obtained with the addition of 1.5 and 2 mL/g of H_2SO_4 . However, $17.57 \pm 2.81\%$ and $15.63 \pm 2.86\%$ for Mn was leached when 1.5 and 2 mL/g of H_2SO_4 was added. Statistically, there was non-significant ($p < 0.05$) difference in leaching efficiency of Cu, Fe and Mn among the different concentrations of H_2SO_4 (Table 1, Figure 1).

Using XRF method, the leaching efficiency of $0.03 \pm 0.01\%$ and $0.04 \pm 0.01\%$ for Cu was obtained with the addition of 1.5 and 2 mL/g of H_2SO_4 . For Fe, leaching efficiencies of $11.13 \pm 2.89\%$ and

$12.80 \pm 3.38\%$ were obtained with the addition of 1.5 and 2 mL/g of H_2SO_4 . However, $0.18 \pm 0.10\%$ and $0.19 \pm 0.09\%$ for Mn was leached when 1.5 and 2 mL/g of H_2SO_4 was added. Statistically, there was non-significant ($p < 0.05$) difference in leaching efficiency of Cu, Fe and Mn among the different concentrations of H_2SO_4 (Table 2, Figure-2).

Table 1. Metal contents of copper tailing after leaching with H_2SO_4 through AAS method.

Technique	Concentration	Cu	Fe	Mn
AAS	1.5 mL	0.99 ± 0.09	13.95 ± 1.42	17.57 ± 2.81
	2 mL	1.00 ± 0.03	11.70 ± 1.22	15.63 ± 2.86
P-value		0.9481	0.2986	0.6548
SE \pm		0.0968	1.8818	4.0157
LSD @ 0.05		0.2686	5.2247	11.149

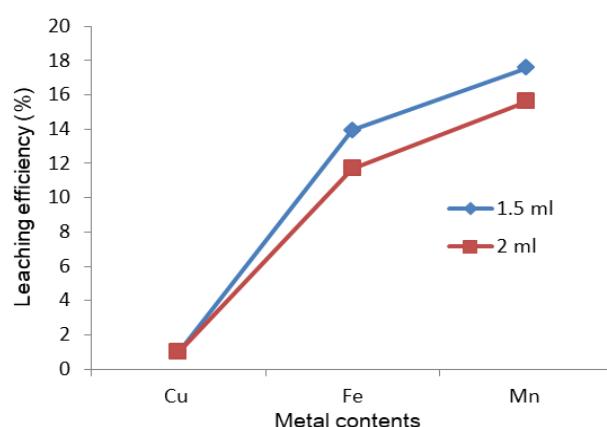


Figure 1. Leaching efficiency of the metal contents using AAS method

Table 2. Metal contents of copper tailing after leaching with H_2SO_4 through XRF method.

Technique	Concentration	Cu	Fe	Mn
XRF	1.5 mL	0.03 ± 0.01	11.13 ± 2.89	0.18 ± 0.10
	2 mL	0.04 ± 0.01	12.80 ± 3.38	0.19 ± 0.09
P-value		0.5072	0.7264	0.9649
SE \pm		0.0137	4.4480	0.1424
LSD @ 0.05		0.0382	12.350	0.3955

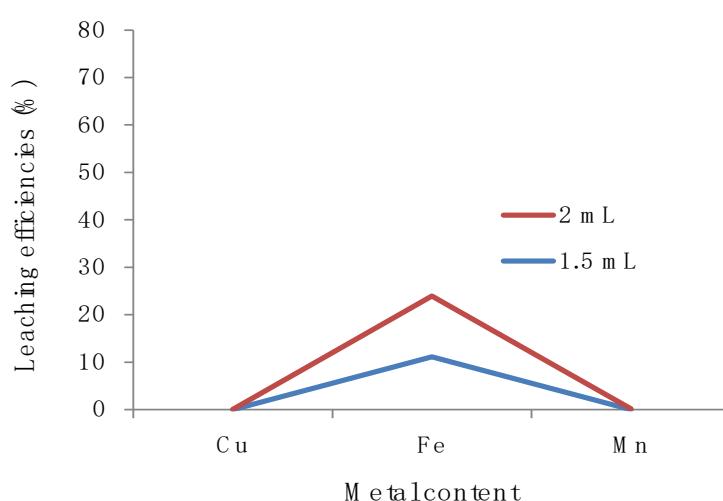


Figure 2. Leaching efficiency of the metal contents using XRF method.

The metal recovery from the leaching filtrate was preceded by fractional precipitation with the addition of different chemical agents. At first, the pH of the leaching filtrate was adjusted to 3.0 with the addition of $\text{Ca}(\text{OH})_2$ to recover Fe as hydroxide. Thereafter, Cu and Zn were recovered as their metal sulfides through the dropwise addition of a 0.5wt % aqueous Na_2S solution in sequence. The pH value of the solution after Cu and Zn precipitations are 3.6 and 3.8 respectively. Finally, Mn was recovered by adjusting the pH to 9.0 in the leaching filtrate. With the UV conversion of the Fe sludge, magnetite particles could be obtained and it can be applied for the paint industry, while the Fe_2O_3 content of the magnetite particles should be higher than 75%.

It has been found that mostly Cu and Zn sludges and those generated in the sulfide precipitation process were covellite and spharelite, and those could be refined and sold to smelters to recover the metals. Generally, the Cu and Zn contents of the sludge are about 0.5% and 2% for their flotation refinement respectively. While the Cu and Zn contents of the sludge were 19.5% and 33.5% respectively. The metal contents of the sludge are higher than these specifications; therefore, the sludge could be refined with the flotation process. Furthermore, Cu and Zn could be obtained through the smelting process of the flotation concentrates. The Mn sludge can be used as cement material and the leaching of heavy metals from the solidified blocks would be negligible.

4. Conclusions

Comprehensive reuse of the tailing would improve economic efficiency and lessen environmental damage as well. Such comprehensive reuse could be promoted by the leaching process. The objective of this work is to recover Cu, Fe, and Mn from low-grade Copper tailing effectively using an environment-friendly and economical leaching method. Cu, Fe and Mn from the low grade copper tailing are successfully recycled via a H_2SO_4 leaching and subsequent fractional precipitation technology. Based on the mineral composition analysis results, leaching temperature, the amount of H_2SO_4 added, leaching time, and S: L have been evaluated for the leaching efficiencies. The amount of H_2SO_4 added and S: L have strongly influenced the leaching efficiencies. Under the optimum conditions of ambient temperature and atmospheric pressure, 0.24mL/g of H_2SO_4 , S: L of 1:2, stirring speed of 400 rpm, and leaching time of 2 h, leaching efficiencies achieved for Cu, Fe and Mn, respectively. With the fractional precipitation process, Fe and Mn were enriched as hydroxide, whereas Cu was enriched as sulfides. The Cu, Fe, and Mn sludges could be further refined and sold to the paint industry or the smelters to recover the metals, and with the recycle disposal, the environment problems caused by the tailing could be relieved. While, the leaching residue reuse needs further consideration for its environment impact.

Acknowledgments

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