COPPER RECOVERY FROM RE-FLOTATION TAILINGS BY COMBINED PROCESS

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Abstract

An efficient method has been proposed to recovering substantial amounts of copper from re-flotation tailings and reducing the environmental impact of flotation tailings by a combined hydrometallurgical process which includes High Pressure Leaching (HPL)-Solvent extraction (SX)-Electrowinning (EW) processes. As a result of HPL using water or sulfuric acid as leaching solution, high copper leaching rate (>95%) was obtained even with water. Then, the copper concentration in the stripped solution reached 44.8 g/L and the iron concentration was 1.4 g/L by the SX tests using LIX-84I as extractant. Moreover, it was confirmed that as lower iron concentration in the electrolyte for the copper electrowinning process, the better the current efficiency of copper electrodeposition.

***Keywords**: copper recovery, flotation tailings, HPL, SX, EW*

1. INTRODUCTION

In the process of copper ore processing, more precisely in the flotation process, which is carried out in order to obtain copper concentrate, flotation tailings are generated as an intermediate product of the process. These tailings are disposed of in a dump, while the copper concentrate is used in the smelting process, after which pure metallic copper is obtained. In the course of long-term mining activities, a large amount of flotation tailings was deposited at the Old Flotation Tailings Dump in Bor, Serbia. Given that the copper content in these tailings is about 0.2-0.4%, there is a possibility that the tailings can be used as a secondary raw material for copper recovery. In order to increase the concentration of copper in the flotation tailings, re-flotation process of the tailings was carried out as a pre-treatment, after which a re-flotation tailings was obtained. In this paper, experimental investigations were carried out on the copper recovery from re-flotation tailings by a combined hydrometallurgical process that includes the three following processes: High Pressure Leaching (HPL) process, Solvent Extraction (SX) process, and Electrowinning (EW) process.

2. EXPERIMENTAL

In order to achieve a high level of copper leaching rate from the re-flotation tailings, which consists of chalcopyrite as the main component of copper, leaching was carried out under high pressure in an autoclave [1]. Then, the solvent extraction of copper from the pregnant leaching solution (PLS) that was obtained by HPL was carried out for obtaining the Cu-rich solution. Finally, a copper electrowinning experiment was carried out in relation to the composition of the solution obtained by applying the optimal process parameters of solvent extraction. In order to perform tests for obtaining copper, it is desirable that the concentration of copper in the electrolyte is high (45 g/L) and the iron concentration is low (under 2 g/L) . Therefore, the influence of iron ions on the current efficiency of copper electrodeposition was confirmed. Current copper electrodeposition efficiency is described in Khouraibchia et al. [2].

2.1 High pressure leaching process

The chemical compositions of re-flotation tailings sample used for HPL tests was as following: Cu 0.34 wt.%, Fe 8.96 wt.%, Al 8.12 wt.%, S 11.2 wt.% and SiO_2 57.8 wt.%. The content of the

investigated elements was determined using Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) and X-ray Fluorescence Spectrometry (XRF). An autoclave device (Nitto Koatsu, Japan) containing a teflon reaction cell with a volume of 2 L was used for the High Pressure Leaching experiments of re-flotation tailings.

The investigated experimental conditions of HPL process were as following: leaching reagents – water or 0.2-0.5 mol/L H_2SO_4, total pressure of O_2 gas 0.8-2.0 MPa, pulp density 100-400 g/L, temperature 140-180 °C and leaching time 60 min.

2.2 Solvent extraction process

In the SX process, the influence of the pH value of PLS obtained after the HPL process on the degree of Cu extraction was investigated. Correction of the pH value was done with a 1.0 M NaOH solution. The following parameters of the SX process were constant: extractant LIX-84I diluted in kerosene in the ratio 1:1, ratio of organic and aqueous phase O/A=0.2, process time 15 min. After the SX process was completed, the solution was centrifuged for 5 min at 4000 rpm in order to rapidly separate the phases.

After determining the optimal conditions of the SX process, the influence of sulfuric acid concentration of 0.5, 1.0 and 1.5 M on the degree of Cu re-extraction process from Cu-rich phase was examined. Other constant parameters of the re-extraction process were as follows: ratio of organic and aqueous phase O/A = 5, duration of the process 15 min.

The concentrations of copper in the aqueous solutions after the extraction and re-extraction processes were determined using ICP-OES, after which the processes efficiencies were calculated.

2.3 Electrowinning process

Conditions of electrowinning process of copper are presented in Table 1. For the EW process, platinum anode and steel cathode with an area of 10x10 mm^2 were used. Before and after the EW process, electrodes were measured and based on the difference in weight, the amount of deposited Cu was recalculated, and also the efficiency of the process.

Table 1 - Experimental conditions of EW process

Volume of electrolyte (L)	Cu concentration (g/L)	Fe concentration (g/L)	Current density (A/m^2)	Temperature (°C)	Free H_2SO_4 (g/L)	Process time (4)
0.5	44.8	1.4	250	40	170	4

3. RESULTS AND DISCUSSION

3.1 High pressure leaching process

Figure 1 presents the influence of H_2SO_4 concentration (0 (water)-0.5 mol/L) on copper recovery from the re-flotation tailings. The copper approximately completely dissolved, while the iron leaching rate reached 95.5%. Even though the sulfuric acid concentration is zero, as leaching reagent water was used, a high degree of copper leaching was achieved. This is due of the generation of sulfuric acid by the oxidation of pyrite as shown in equation (1) [4]. As a result of the formation of sulfuric acid, there was a decrease in the pH value of the slurry from 3.79 to 0.35 after the HPL process using water as leaching reagent.

$$2FeS_2 + 7.5O_2 + H_2O \rightarrow Fe_2(SO_4)_3 + H_2SO_4 \quad (1)$$

After experimental investigations of the HPL process, it can be concluded that the optimal process parameters are as follows: water as leaching reagents, total pressure of O_2 gas: 2.0 MPa, pulp density: 100 g/L, temperature: 180 °C and leaching time: 60 min.

From an economic point of view, water is selected to be the leaching reagent for copper leaching from the re-flotation tailings. Under the optimal HPL conditions, leaching rate of Cu is 94.4% and the Cu concentration in the obtained PLS is 2.9 g/L.
To further recovery of copper from PLS, a solvent extraction process was carried out using the leaching solution obtained under the optimal HPL conditions.

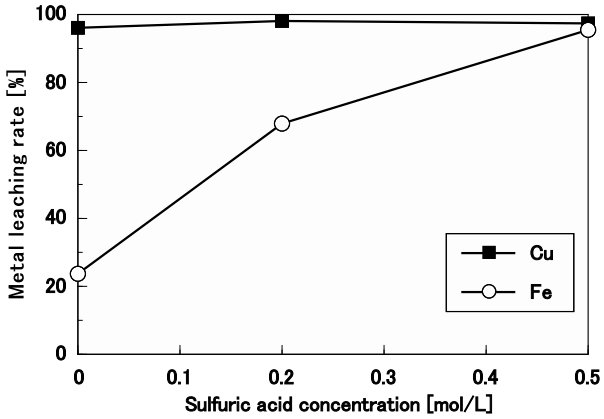

Figure 1 - Leaching rate of copper and iron as a function of sulfuric acid concentration (Total pressure: 2.0 MPa, temperature: 180 °C, pulp density: 100 g/L, leaching time: 60 min)

3.2 Solvent extraction process

The results of the study of the influence of the pH value of the PLS on the EX process show that the degree of copper extraction depends on the equilibrium pH values that minimize the extraction of other metal ions such as iron [1]. For the EW process, for the selective extraction of copper, the pH of the PLS of 2 was determined as the optimal value.

As for the re-extraction process, the degree of copper recovery from the Cu-rich organic phase increases linearly from 24.7 to 97.4% with increasing H_2SO_4 concentration from 0.5 to 1.5 M. A 1.5 M solution of H_2SO_4 was chosen as the optimal parameter for the copper re-extraction process from the Cu-rich organic phase. The concentration of copper and iron in the solution obtained after re-extraction contains approximately 44.8 g/L Cu and 1.4 g/L Fe.

3.3 Electrowinning process

The results obtained for the process of electrochemical extraction of copper from the solution - EW process, indicated a high level of current efficiency, over 98%, with the following parameters: copper concentration in the electrolyte 44.8 g/L, H_2SO_4 concentration 170 g/L, current density 250 A/m^2, electrolyte temperature 40^{o}C and time 4 h. The concentration of iron in the electrolyte is limited to a value below 2 g/L. Based on the examination of the influence of the presence of Fe ions in the electrolyte, it was determined that the best current efficiency was achieved in the presence of only ferrous (Fe^{2+}) ions in the electrolyte. The presence of ferric (Fe^{3+}) ions in the electrolyte leads to reduced current efficiency.

4. CONCLUSION

An effective method for copper recovery from re-floation tailings and reducing the negative impact that flotation tailings have on the environment is investigated. The process consists of a combination of the following processes: high pressure leaching (HPL) - solvent extraction (SX) – electrowinning (EW) process.

After the HPL process using water as a leaching reagent under optimal conditions (total pressure of O_2 gas: 2.0 MPa, pulp density: 100 g/L, temperature: 180°C and leaching time: 60 min) a copper leaching rate of >95% was achieved. In the SX process, which had an aim to concentrate Cu and

purify the pregnant leaching solution from Fe under optimal conditions (extraction process: pH of the solution 2, extractant LIX-84I diluted in kerosene in the ratio 1:1, O/A=0.2, time 15 min; re-extraction process: concentration of H_2SO_4 1.5 M, O/A = 5, time 15 min), the concentration of Cu is 44.8 g/L, and Fe 1.4 g/L. The current efficiency of copper electrodeposition in the EW process decreases with increasing iron ion concentration.

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