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Copper Recovery and Reduction of Environmental Loading from Mine Tailings by High-Pressure Leaching and SX-EW Process

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Abstract: The flotation tailings obtained from Bor Copper Mine contain pyrite (FeS_2) and chalcopyrite ($CuFeS_2$), these sulfide minerals are known to promote acid mine drainage (AMD) which poses a serious threat to the environment and human health. This study focuses on the treatment of mine tailings to convert the AMD supporting minerals to more stable forms, while simultaneously valorizing the mine tailings. A combination of hydrometallurgical processes of high-pressure oxidative leaching (HPOL), solvent extraction (SX), and electrowinning (EW) were utilized to recover copper from mine tailings which contain about 0.3% Cu content. The HPOL process yielded a high copper leaching rate of 94.4% when water was used as a leaching medium. The copper leaching kinetics were promoted by the generation of sulfuric acid due to pyrite oxidation. It was also confirmed that a low iron concentration (1.4 g/L) and a high copper concentration (44.8 g/L) obtained in the stripped solution resulted in an improved copper electrodeposition current efficiency during copper electrowinning. Moreover, pyrite, which is primarily in the mine tailings, was converted into hematite after HPOL. A stability evaluation of the solid residue confirmed almost no elution of metal ions, confirming the reduced environmental loading of mine tailings through re-processing.

Keywords: tailings valorization; high-pressure leaching; copper recovery; acid mine drainage; metal elution

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1. Introduction

Mine tailings are waste material generated from the flotation process and there are billions of tons of already existing tailings all over the world. Moreover, several billion tons of additional mine tailings will inevitably be produced, as lower-grade and complex ores are being mined to sustain the world's growing demand for mineral resources [1,2]. Schlesinger et al. [3] (p.68) estimated that flotation tailings account for 98% of the ore fed in to concentrators, which are stored in large tailings dams near their mines. Unfortunately, the storage of mine tailings is associated with severe environmental challenges. If not managed properly, the failure of mine tailings storage facilities can result in catastrophic ramifications and environmental pollution, such as the well-known Brumadinho and Mariana tailings dam disasters in Brazil. The most serious environmental impact resulting from mine tailings is the generation of acid mine drainage (AMD). Mine tailings that contain metal sulfides, such as pyrite (FeS_2), are a major source of AMD [4–6]. Pyrite is a gangue mineral that is commonly found in mine tailings, when exposed to an environment with oxygen and water, oxidation of pyrite is promoted via reaction 1, which results in low pH conditions of less than 3 [5,7–9].

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

The low pH conditions are detrimental because they increase the dissolution and mobility of toxic and heavy metals/metalloids such as Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Zn, As, and Se [6,10]. When these toxic metals accumulate in the environment, they become a major source of contamination to water sources and potentially endanger ecosystems and human health [5,6,8,11]. The flotation tailings from Bor Copper Mine contain pyrite and chalcopyrite, therefore, management of these mine tailings is extremely important, as well as developing effective long-term strategies to reduce the environmental footprint of the mining industry.

The reprocessing of mine tailings is now being globally explored as an approach to be sustainable in both active and inactive mining operations. As new technologies emerge, valuable metals can efficiently be extracted from mine tailings with reduced environmental impacts. Most old copper mine tailings have a comparable copper grade to that of low-grade ores (0.2–0.3%) and considering their vast amount, they present a potential source of secondary raw material for copper production [6,7,12–15]. Several technologies have been used for the treatment of mine tailings to extract valuable metals, these mostly include bioleaching [12,15–17], flotation [10,14,18], hydrometallurgical methods [4,6,7,19], or a combination of these treatment methods [13,20]. Hydrometallurgical methods are commonly considered for the treatment of low-grade sulfide material because of comprehensive metal recovery and shorter retention times. Antonijevi'c et al. [7] investigated the leaching of flotation tailings from Bor Copper Mine using sulfuric acid and iron (III) sulfate as an oxidant and obtained a copper recovery of 80% after 2 h, highlighting that the flotation tailings should be valorized rather than used for land reclamation. Chen et al. [6] demonstrated that low-grade copper sulfide tailings with pyrite proportions of 31.50%, can be recycled for environmentally friendly disposal through leaching and fractional precipitation methodology, from which a copper recovery of 98.45% could be achieved. Selective extraction of copper from tailings was successively investigated by Turan et al. [21] using a mixture of ammonia salts as leaching reactants and managed to obtain a copper recovery of 91.47% with no iron tenors to the leachate after 6 h of leaching at 30 °C.

This study evaluates the high-pressure oxidative leaching (HPOL) method as an effective way to recover copper and reduce the environmental loading from the mine tailings obtained from Bor Copper Mine, Serbia. The advantages of pressure leaching include fast kinetics, enhanced metal selectivity, and the generation of stable residues [22]. Han et al. [23] have already presented in full detail about high-pressure leaching of the concentrate obtained after the flotation of the mine tailings. This study considers high-pressure leaching directly on the mine tailings as well as environmental evaluation of the leach residue stability. Particular interest was given to the high-temperature oxidation treatment of the mine tailings to convert the reactive and harmful minerals such as pyrite, to more stable forms such as hematite (Fe_2O_3). Chalcopyrite ($CuFeS_2$) has also been listed as one of the sulfide minerals important in AMD formation [5]. Therefore, alongside the conversion of pyrite to hematite, the dissolution of residual chalcopyrite from flotation tailings will also aid in the prevention of AMD. To evaluate the effectiveness of this method in reducing the environmental loading caused by flotation tailings, metal elution tests were performed on the solid residue obtained after HPOL and on the flotation tailings. In addition to Han et al.'s [23] study, this research further investigates the electrowinning process for final copper recovery, in order to demonstrate the feasibility of a complete hydrometallurgical process when utilizing the discarded mine tailings. The electrowinning process was carried out to examine the effect of iron and copper concentration of the stripped solution on current efficiency for copper electrodeposition.

2. Materials and Methods

2.1. Experimental Sample

The mine tailings sample used in this study was obtained from Bor Mine in Serbia. The sample was obtained 14 m below the ground surface of the tailings dam. The average particle size of the mine tailings sample was 27 μm (D_{50}). Table 1 shows the chemical compositions of the two samples utilized in this study; the mine tailings and the concentrate obtained after the flotation of the mine tailings. X-ray diffraction (XRD) analysis (Figure 1) showed that the main mineral compositions of the mine tailings and concentrate of the mine tailings were quartz (SiO_2), pyrite (FeS_2), and kaolinite ($Al_2Si_2O_5(OH_4)$). The main copper mineral in the mine tailings was identified as chalcopyrite ($CuFeS_2$) using scanning electron microscope-energy dispersion spectroscopy (SEM-EDS) (Figure 2). The copper concentrate used in this study was prepared by flotation under determined optimal conditions: pH 10, pulp density 25%, PAX collector dosage 100 g/t-ore, NaHS sulfidizer dosage 1000 g/t-ore, MIBC frother dosage 200 g/t, flotation time 5 min [24]. The obtained copper concentrate and the original mine tailings were both used as feed samples in the subsequent leaching experiments.

Table 1. Chemical compositions of the mine tailings and concentrate.

	Grade (mass%)				
Elements	**Cu**	**Fe**	**Al**	**S**	**SiO_2**
Mine tailings	0.24	3.51	3.45	4.88	61.7
Concentrate from mine tailings	0.65	33.20	2.63	32.72	23.41

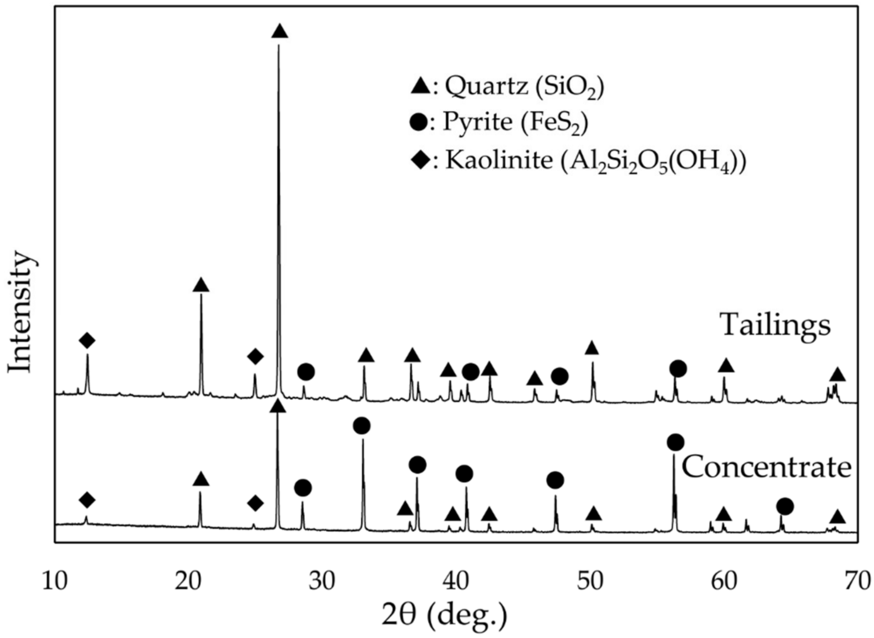


Figure 1. XRD patterns of mine tailings from Bor Copper Mine and concentrate of mine tailings.

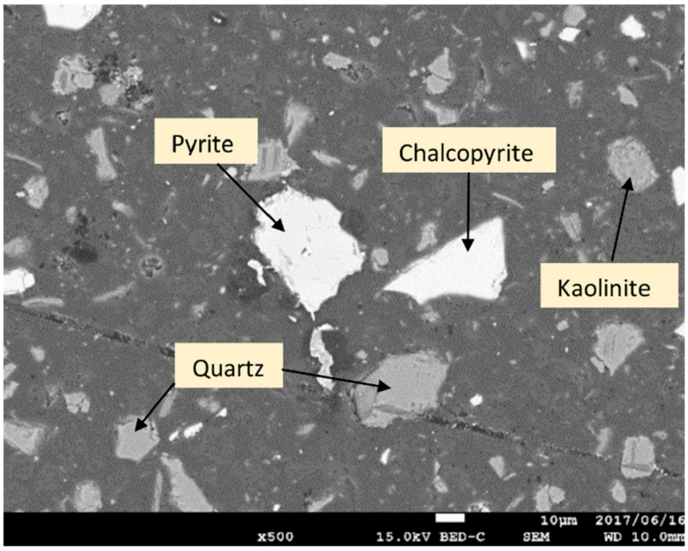


Figure 2. SEM image of the mine tailings from Bor Copper Mine.

2.2. Experimental Procedure

2.2.1. High-Pressure Leaching

An autoclave of a 200 mL Teflon beaker was used as a leaching reactor in the high-pressure oxidative leaching experiment (Figure 3) for the mine tailings and concentrate from mine tailings. A sample weight of 10 g was mixed with 0–1.0 M sulfuric acid (H_2SO_4) solution, where 0 M was distilled water, to obtain a pulp density of 100 g/L. The slurry was then placed inside the autoclave and heated. After the temperature reached 180 °C, oxygen gas was injected at 2 MPa into the slurry to provide an oxidative environment. The leaching duration was set to 1 h. After the HPOL experiment, the sample was cooled and filtered to obtain a pregnant leach solution (PLS) and a solid residue, which were taken for chemical and mineralogical analysis using ICP-OES and XRD, respectively.

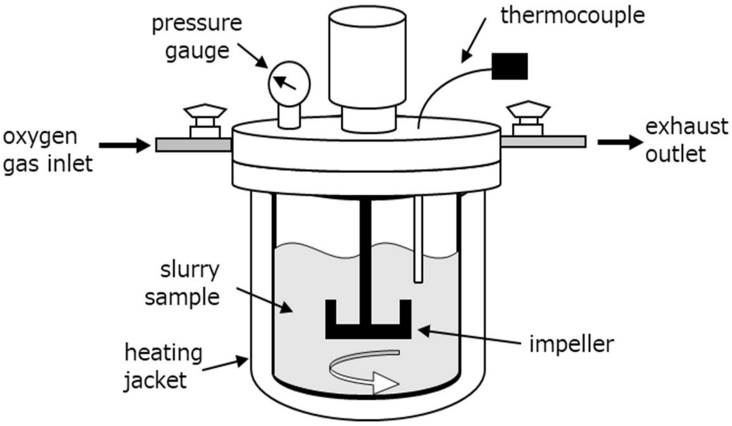


Figure 3. Illustrative setup of an autoclave reactor used for high-pressure oxidative acid leaching.

2.2.2. Elution Test

Elution properties of the mine tailings and the solid residue obtained after HPOL were evaluated. A sample weight of 0.4 g was placed in a sample tube (capacity: 5 mL) together with 4 mL of different pH solutions (pH 2 (adjusted with H_2SO_4), pH 4 (distilled water), and pH 7 (adjusted with $Ca(OH)_2$)). The solutions were shaken for 6 h at 200 rpm, using a shaker (MMS-4020, EYELA). Afterward, the slurry was subjected to solid-liquid separation by centrifugation. Quantitative analysis of each metal in the solution was carried out using ICP-OES (SPS5500, SII nanotechnology). The elution rate was calculated as per Equation (2).

$$\text{Elution rate}(\%) = \frac{C_S * V_S}{C_R * m_R} * 100 \tag{2}$$

where C_S is the concentration of metal in the solution obtained from the elution test (mg/L), and C_R is the concentration of metal in the sample (leach residue/mine tailings), (mg/kg). V_S is the volume of solution (L) and m_R is the dry mass of the sample used (kg).

2.2.3. Solvent Extraction Test

Extraction of copper from the PLS obtained after HPOL of the concentrate from mine tailings was previously carried out by Han et al. [23] using LIX-84I (2-hydroxy-5-nonylace-tophenone oxime) extractant. Kerosene was used to dilute the extractant at a phase ratio of 1. A 10 mL of the PLS and 2 mL of diluted extractant (organic/aqueous phase ratio = 0.2) were placed in a plastic tube and the pH was adjusted to the desired value using a 1.0 M NaOH solution. The solution was mixed using an agitator at 600 rpm and the extraction time was set to 15 min. Afterward, the solution was centrifuged for 5 min at 4000 rpm to ensure a rapid phase separation. To determine the metal extraction efficiency, the raffinate was analyzed using ICP-OES. The Cu-loaded organic obtained from the optimal copper extraction conditions was subjected to stripping experiments where different sulfuric acid concentrations (0.5, 1.0, and 1.5 M) were mixed with the Cu-loaded organic (organic/aqueous phase ratio = 5) at 600 rpm and 15 min contacting time. The stripped solution was analyzed by ICP-OES for the determination of the stripping efficiency and copper concentration [23].

2.2.4. Electrowinning Test

Electrowinning of copper was carried out with a simulated solution prepared in consideration of the copper and ferric ion concentrations of the stripped solution from the solvent extraction stage (44.8 g/L Cu and 1.4 g/L Fe) using distilled water, copper sulfate (II) pentahydrate ($CuSO_4{\cdot}5H_2O$) and iron (III) sulfate n-hydrate ($Fe_2(SO_4)_3{\cdot}nH_2O$), where n was analyzed to be 6.33. The electrolyte also contained 170 g/L of free $H_2SO_{4,}$ and no additives were used. The electrolyte temperature was maintained at 40 °C with a bath heater and the volume of the electrolyte was 0.5 L. The anode and cathode were made from a platinum plate of 1 cm^2 surface area and mounted on epoxy resin with a conducting wire. The anode and cathode were positioned in a fixed mount facing each other, the distance between the electrodes was 3 cm and the mount was placed in the preheated electrolyte cell. A regulated DC power supply (Takasago, Ltd. GP025-5) was used, the operating current density was set at 250 A/m^2, and the electrolysis time was 4 h. To further study the effects of metal concentration in the electrolyte, the copper and iron concentrations were varied between 25–45 g/L and 0–1.5 g/L, respectively. The cathode was weighed before and after the electrolysis test to determine the weight of copper plated. The current efficiency was calculated as per Equation (3). The theoretical mass of the copper deposited was calculated using Equation (4):

$$Current\ Efficiency,\ (C.E) = \frac{mass\ of\ Cu\ deposited\ (actual)}{mass\ of\ Cu\ deposited\ (theoretical)} * 100 \tag{3}$$

$$Cu\ deposited\ (g) = \frac{mm * j * A * t}{nF} \tag{4}$$

where mm is the molar mass of copper, j is the current density, A is the electrode surface area, t is the electrolysis time, n is the number of electrons and F is faradays constant.

3. Results and Discussion

3.1. High-Pressure Leaching

3.1.1. Leaching of Mine Tailing

The effect of sulfuric acid concentration on direct pressure leaching of mine tailings was investigated using distilled water and 0.2–1.0 M H_2SO_4. Other leaching conditions were kept the same as in Han et.al [23] at a fixed pulp density of 100 g/L, total pressure of 2 MPa, leaching temperature of 180 °C, and leaching time of 1 h. As shown in Figure 4, the highest achieved leaching rate of Cu was 98.72% when distilled water was used as a leaching medium, while the lowest achieved leaching rate of Fe was 16.31% at the same leaching conditions. The Cu and Fe concentrations in the leachate were 0.23 g/L and 0.28 g/L, respectively. The high leaching rate of Cu can be attributed to the oxidation of pyrite which is one of the main minerals in the mine tailings. The pyrite was oxidized as per reaction 1 under a total pressure of 2 MPa and leaching temperature of 180 °C, thus generating sulfuric acid [7,9,25]. This was verified by a decline in the pH value from 3.12 before leaching to 0.8 after leaching. The sulfuric acid generated by pyrite oxidation promoted the leaching of chalcopyrite via reaction 5. Han et al. [23] confirmed that the presence of pyrite (FeS_2) in the feed has an efficient effect on copper dissolution. Antonijevic et al. [7] also found that with the increasing concentration of H^+ ions in the leach solution, the dissolution of copper increased. Figure 5 shows the Eh–pH diagram, where the point indicates the pH and Eh (pH: 0.8, Eh: 676 mV) of the solution obtained after leaching. The stability of Cu^{2+} in solution is favored by the leaching conditions of a high temperature and high pressure thus enabling a high leaching rate. XRD measurement of the solid residue (Figure 6) showed the presence of hematite (Fe_2O_3) and the absence of pyrite which was initially in the mine tailings. This indicates that the majority of ferric sulfate is hydrolyzed at a high temperature and total pressure to form hematite as shown by reaction 6. The precipitation of Fe as hematite resulted in a high metal selectivity of dissolved copper over iron when only water was used as a leaching medium.

$$2CuFeS_2 + H_2SO_4 + 8.5O_2 \rightarrow 2CuSO_4 + Fe_2(SO_4)_3 + H_2O \quad (5)$$

$$Fe_2(SO_4)_3 + 3H_2O \rightarrow Fe_2O_3 + 3H_2SO_4 \quad (6)$$

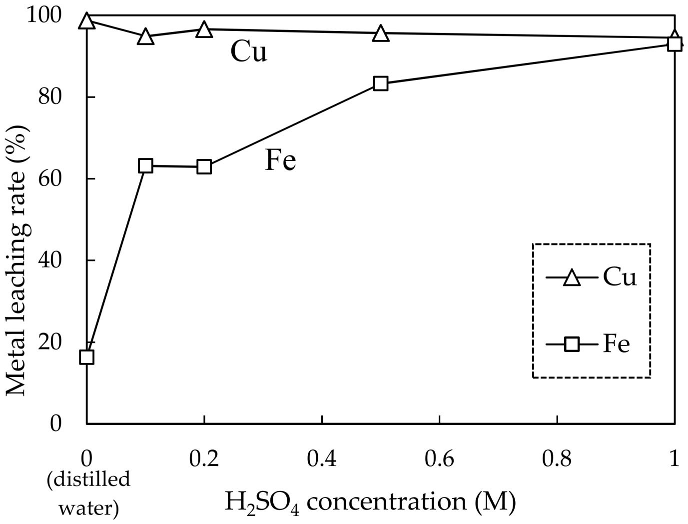


Figure 4. Effect of sulfuric acid concentration on the leaching rate of copper and iron. (Conditions: 100 g/L, 1 h, 700 rpm, 180 °C, 2.0 MPa total pressure).

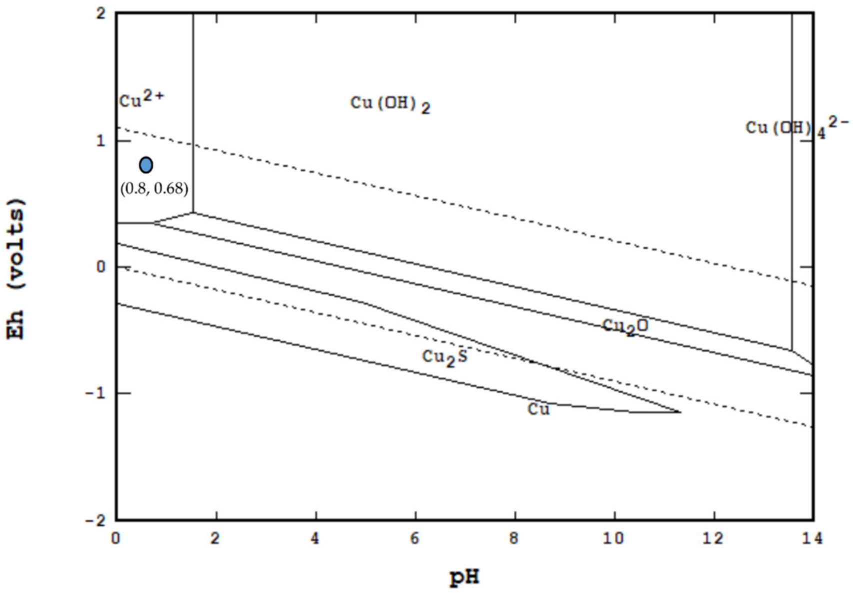


Figure 5. Eh−ph diagram for Cu−S−water system calculated by STABCAL (Condition: 180 °C, 2 MPa).

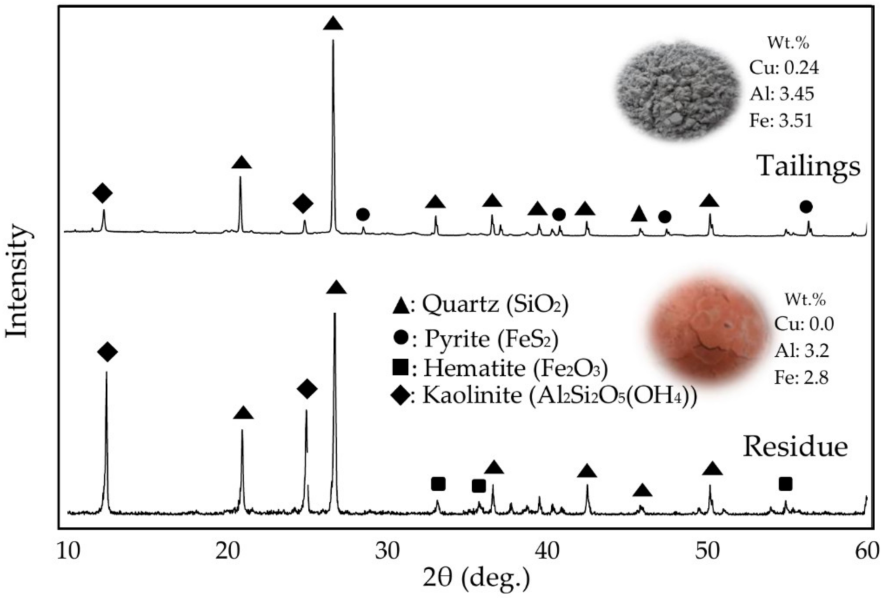
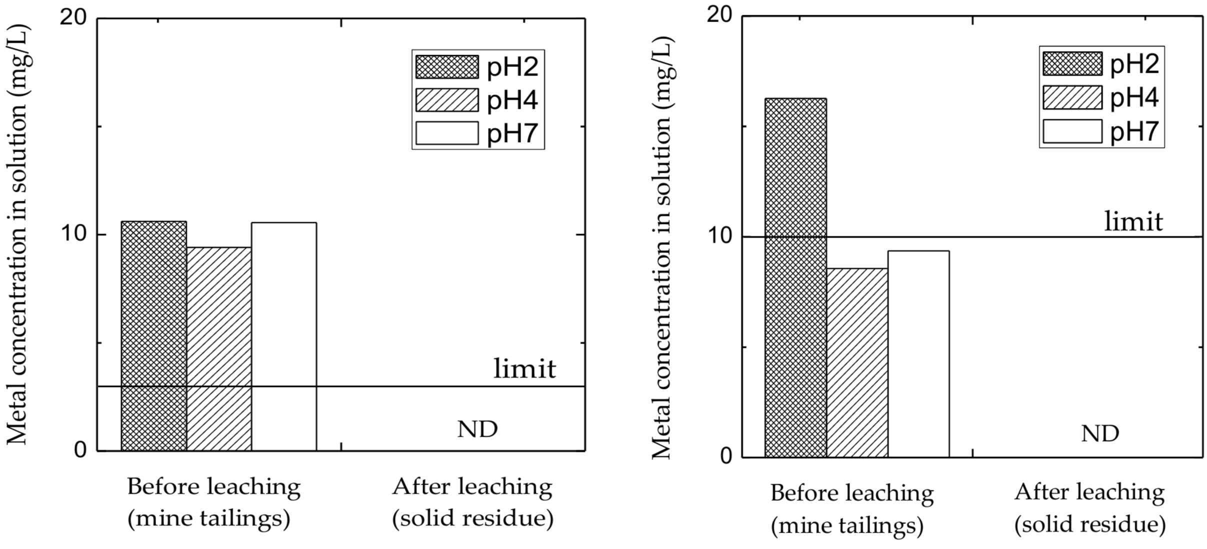


Figure 6. XRD patterns of the residue obtained after leaching of mine tailings with distilled water and the mine tailings.

3.1.2. Elution Test of Leaching Residue Obtained from High-Pressure Leaching

The elution test results are displayed in Figure 7a,b, showing copper and iron concentrations in the solution, respectively. The national effluent standards of copper and iron in Serbia could not be identified, therefore Japan was used for comparison of the results obtained from the elution test. Based on the wastewater discharge standards specified by the Ministry of the Environment in Japan the upper limit for Cu and Fe in the wastewater discharge is 3 mg/L and 10 mg/L, respectively [26]. The concentration of copper in the solution of mine tailings exceeded this criterion at all the investigated pH values (Figure 7a), while the iron concentration in solution exceeded this criterion at a low pH value of 2 (Figure 7b). The elution rate of copper and iron from the mine tailings was calculated to be 4.58% and 0.46% respectively. On the other hand, the solid residue obtained from leaching the mine tailings could meet the regulatory metal criterion with a very low metal concentration below the detection limit (<0.1 ppm) of ICP-EOS. Under various pH conditions, Fe did not elude, mainly because hematite is thermodynamically stable and therefore, less soluble. Furthermore, the elution rate of copper in the solid residue remained undetectable because 98.72% of copper has been recovered in the PLS. It can thus be confirmed that the elution rate of the mine tailings can be reduced by the HPL process, generating a benign solid residue of a reduced environmental loading.



(**a**) (**b**)

Figure 7. Metal concentration in the solution obtained from the elution test of mine tailings (before leaching) and solid residue (after leaching): (**a**) copper; (**b**) iron. ND = not detected.

3.1.3. Leaching of Concentrate from Mine Tailings

The grade of copper in mine tailings was upgraded from 0.24% to 0.65% through flotation. The concentrate was subjected to high-pressure leaching under the optimal conditions obtained from leaching the mine tailings (leaching medium distilled water, leaching time 1 h and total pressure 2 MPa controlled by O_2 gas, temperature 180 °C) [23]. To concentrate copper by solvent extraction and obtain a solution suitable for electrowinning, a higher copper concentration in the PLS than that obtained from high-pressure leaching of mine tailings (0.23 g/L) is necessary. For this reason, the pulp density was adjusted to 400 g/L. Muravyov and Fomchenko [20], Antonijevic et al. [7], and Muravyov et al., [4] found that the pulp density has no significant effect on the leaching of metals from flotation tailings after obtaining similar values of dissolution degree with various pulp densities. Han et al. [23] achieved a high copper leaching rate of 94.4%, showing no significant change when compared to the leaching rate of Cu from mine tailings (98.72%) at a lower pulp density of 100 g/L. However, increasing the pulp density to 400 g/L increased the leaching rate of iron up to 65.9%. This is attributed to the increase in the added amount of pyrite, which resulted in an increase in free acid concentration in the solution and thus an increase in iron solubility [23]. The obtained PLS consisted of 2.9 g/L of Cu and 102.9 g/L of Fe with a pH of 0.23. For further recovery of copper from the PLS, solvent extraction was carried out.

3.2. Solvent Extraction of the Pregnant Leach Solution from HPOL

Han et al. [23] reported on the results of the extraction of copper from the PLS under the optimal HPOL conditions and extraction conditions stipulated in Section 2.2.3. Figure 8 was drawn based on the results obtained from Han et al. [23], which show that the extraction of copper increased from 10.1 to 93.7% as the pH was increased. However, the iron extraction rate remained below 11.3% yielding a good separation efficiency of 82.4% between copper and iron. The pH of 2.0 was established as an optimal pH for the selective extraction of copper from the PLS.

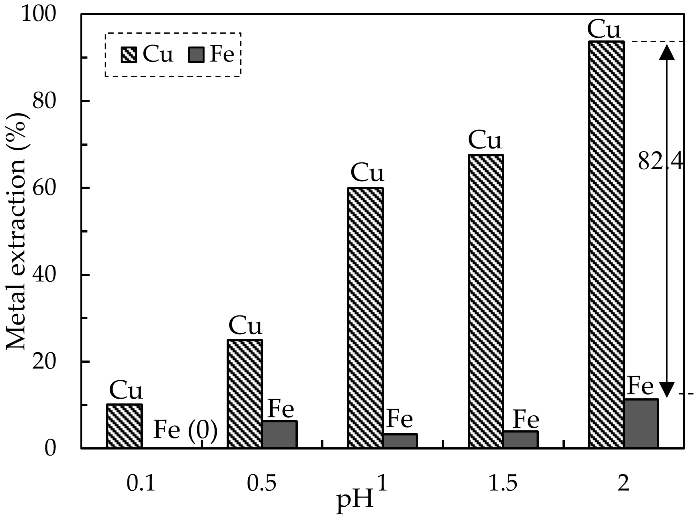


Figure 8. The extraction behavior of metals as a function of pH (Conditions: LIX-84I/Kerosene mixing ratio 1, organic/aqueous phase ratio 0.2, agitating speed 600 rpm, and contact time 15 min).

Han et al. [23] also examined the stripping efficiency of copper from the Cu-loaded organic phase using three different H_2SO_4 concentrations of 0.5, 1.0, and 1.5 M. The highest copper stripping efficiency of 97.4% was obtained at an H_2SO_4 concentration of 1.5 M and was selected as the most suitable H_2SO_4 concentration for stripping copper from the Cu-loaded organic phase. Figure 9 was prepared considering the data from Han et al. [23] of the metal concentrations in stripped solution as a function of H_2SO_4 concentration. As the H_2SO_4 concentration was increased, the copper concentration in the stripped solution increased while the iron concentration decreased. The optimal stripped solution contained approximately 44.8 g/L copper and 1.4 g/L iron [23]. This is comparable to the industrial electrolyte from the solvent extraction stage which typically contains 45 g/L Cu and less than 2.0 g/L of Fe concentration [3] (p. 356).

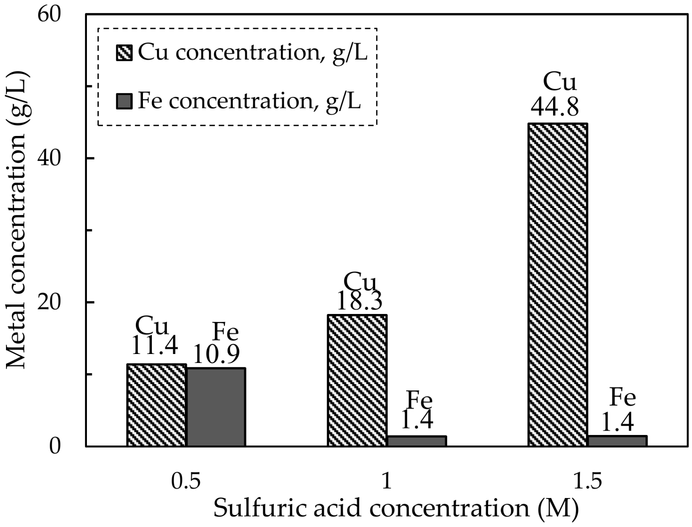


Figure 9. The metal concentration in stripped solution as a function of sulfuric acid concentration. (Conditions: organic/aqueous phase ratio 5, agitation speed 600 rpm, and contact time 15 min).

3.3. Electrowinning of the Simulated Stripped Solution

Current efficiency is an important parameter in electrowinning as it indicates the effectiveness of the electrowinning process. Ferric ion concentration is known to have negative interactions that significantly affect the current efficiency of copper electrodeposition during electrowinning. Therefore, to evaluate the efficiency of carrying out the electrowinning process on the obtained stripped copper solution, it is relevant to examine the current efficiency of the electrowinning process due to the inevitable Fe contamination during leaching. Figure 10 shows the influence of ferric ion concentration on the current efficiency of copper electrodeposition. As expected, a strong inverse relationship is observed, as the ferric ion concentration increases, the current efficiency of copper electrodeposition decreases. This is due to the presence of iron in the copper electrolyte which undergoes reduction from Fe^{3+} to Fe^{2+} at the cathode and re-oxidation to Fe^{3+} from Fe^{2+} at the anode. The cycle proceeds and consumes power that could be used for the deposition of copper. Considering the obtained Fe concentration of 1.4 g/L in the stripped solution, the expected current efficiency is approximately 95%, which is in the upper limit of the industrial range of 85–95% [3] (p. 358). Current efficiency loss was determined to be 2.8% per g/L of ferric ions, which agrees with Khouraibchia and Moats [27] 's empirical model of current efficiency (Equation (7)) and Schlesinger et al., [3] (p. 361)'s findings that current efficiency drops by approximately 2.5% for each addition of 1 g/L of ferric ions.

$$\begin{aligned}\text{Current efficiency}\quad & (\text{Khouraibchia and Moats 's empirical model}) \\ & = 88.19 - 4.19 * [\mathrm{Fe}^{3+}](\mathrm{g/L}) + 0.52 * [\mathrm{Cu}^{2+}](\mathrm{g/L}) \\ & + 1.81 * 10^{-3} * \mathrm{j}(\mathrm{A/m}^2) - 6.83 * 10^{-3} * \left[\mathrm{Cu}^{2+}\right]^2(\mathrm{g/L}) \\ & + 0.028 * [\mathrm{Fe}^{3+}](\mathrm{g/L}) * \left[\mathrm{Cu}^{2+}\right](\mathrm{g/L}) + 4.015 * 10^{-3} \\ & * [\mathrm{Fe}^{3+}](\mathrm{g/L}) * \mathrm{j}\,(\mathrm{A/m}^2)\end{aligned} \tag{7}$$

where j is the current density.

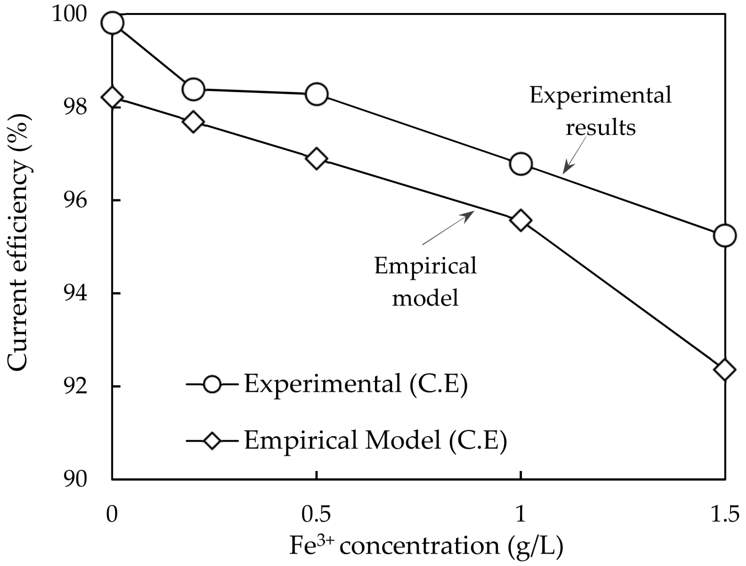


Figure 10. Effect of Fe^{3+} on current efficiency (C.E) of copper deposition. (Conditions: current density 250 A/m^2, temperature 40 °C, electrolysis time 4 h, Cu^{2+} concentration 45 g/L).

Alongside the targeting of a low Fe concentration in the stripped solution, enriching copper concentration was also an important factor during the extraction process. To demonstrate the importance of obtaining a high Cu^{2+} concentration in the stripped solution, the effect of three copper concentrations (25, 35, and 45 g/L) on current efficiency during electrowinning was investigated. The Fe^{3+} concentration in the electrolyte was varied from

0 to 1.5 g/L for each copper concentration investigated. Figure 11 displays the results obtained, showing that current efficiency losses are higher on dilute copper solutions than on concentrated copper solutions. From the gradients of the three plots, current efficiency loss per g/L of Fe^{3+} was determined and is shown in Table 2. A solution of 45 g/L copper concentration had the lowest current efficiency loss. This could be explained by the way that a high Cu^{2+} concentration in the electrolyte constantly provides sufficient copper ions to the cathode surface, thus improving deposition rate as well as the copper current efficiency [28]. Das and Krishna [29] also wrote that increasing the bath Cu^{2+} concentration increases the electrolyte viscosity, which impedes the distribution of Fe^{3+} over the cathode surface. Therefore, in this investigation, obtaining a high copper concentration of 44.8 g/L significantly contributed to achieving a good current efficiency of about 95%.

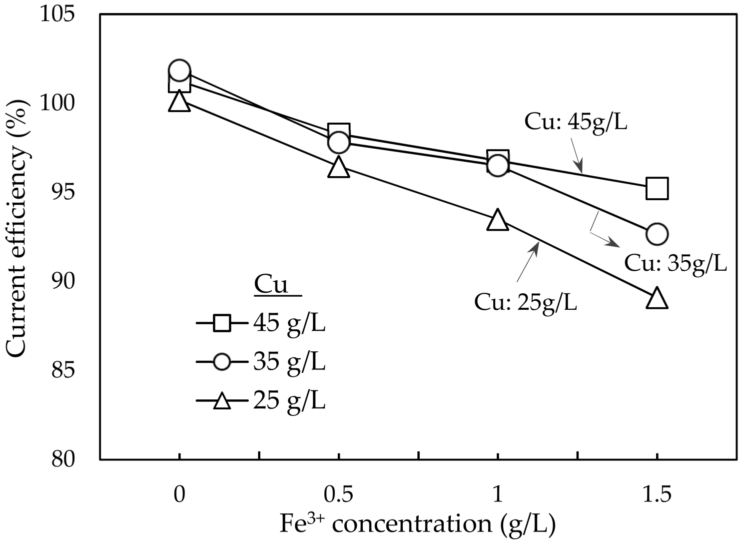


Figure 11. Effect of Fe^{3+} and Cu^{2+} concentration on Cu current efficiency. (Conditions: current density 250 A/m^2, temperature 40 °C, electrolysis time 4 h).

Table 2. The effect of copper concentration on current efficiency loss per g/L of Fe after the electrowinning experiments.

Cu^{2+} Concentration (g/L)	Current Efficiency Losses (Loss per g/L Fe (%))
25	3.62
35	2.88
45	1.95

In hydrometallurgical processes like these, where pyrite is vital to the leaching stage, an Fe contamination in the stripped solution is inevitable. Therefore, obtaining a high copper concentration in the stripped solution is important as it is less susceptible to current efficiency losses during electrowinning. Since the electrowinning experiments were conducted using synthetic solutions, the expected current efficiency on the real solution obtained from solvent extraction may be slightly lower than that obtained when utilizing a synthetic solution. This is due to the complexity of the stripped solution composition because of several other impurities that may cause side reactions. However, current efficiency loss due to ferric ions reduction has been found to have a significant effect on current efficiency compared to other impurities which showed no effect [30]. Therefore, the results obtained from the electrowinning of a simulated solution can be highly comparable to those obtained from electrowinning the real stripped solution under optimized conditions.

4. Conclusions

This study demonstrated that leaching mine tailings could alleviate the possible environmental effects they can pose if left untreated, while simultaneously valorizing them. As an AMD preventative technique, the removal of pyrite from mine tailings through oxidative high-pressure leaching was successively achieved, resulting in the formation of hematite in the solid residue. The obtained residue from leaching the mine tailings, and the original sample of the mine tailings, were subjected to standardized elution tests. The eluded metal concentrations for the solid residue could meet the Ministry of the Environment of Japan's set limits for copper and iron in the wastewater discharge, while the original mine tailings exceeded the set limits, thus confirming the reduced environmental loading of the proposed copper recovery process.

A viable copper recovery process for re-treatment of mine tailings was developed by employing hydrometallurgical methods of high-pressure leaching, solvent extraction, and electrowinning. During high-pressure leaching, the generation of sulfuric acid due to pyrite oxidation promoted copper leaching kinetics when water was used as a leaching medium, thus yielding a high copper leaching rate of 94.4%. Under the optimized solvent extraction conditions, over 93.7% of copper was extracted from the PLS while most of the iron was left in the organic phase. A high copper stripping efficiency of 97.4% was obtained resulting in an enriched solution containing 44.8 g/L Cu and 1.4 g/L Fe. Due to a minimized iron carryover from the stripped solution, electrowinning power consumption by iron was greatly reduced and current efficiency for copper electrodeposition was over 95%.

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