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# LEACHING OF COVELLITE IN IONIC LIQUID SOLUTION

**Dragana Marilović[1a], Grozdanka Bogdanović[1b]**

[1] Technical Faculty Bor, University of Belgrade, V.J. 12, 19210 Bor, Serbia
[1a] dmarilovic@tfbor.bg.ac.rs, 0000-0001-8301-750X
[1b] gbogdanovic@tfbor.bg.ac.rs, 0000-0002-1013-4075

**Abstract**
*Covellite is refractory to conventional hydrometallurgical processes, therefore alternative methods are required to improve copper extraction. The aim of this work was to investigate the leaching of covellite concentrate in an ionic liquid solution ([bmim]$HSO_4$) with hydrogen peroxide as a strong oxidizing agent. The results indicate that the presence of hydrogen peroxide significantly improves copper extraction due to its strong oxidizing ability, which facilitates the dissolution of sulfide species. Although only preliminary data are presented, the results confirm the potential of ionic liquids in combination with oxidizing agents for the treatment of copper sulfide concentrates. When covellite concentrate was leached in an ionic liquid solution without oxidants, the leaching degree of copper was 1.03 %. The results showed that the maximum dissolution of copper in the presence of 3.0 mol/dm$^3$ hydrogen peroxide was 28.97 %. This approach provides a basis for the development of more environmentally friendly and efficient leaching processes for complex copper ores and concentrates.*

***Keywords**: Covellite, Ionic liquid, Hydrogen peroxide, Leaching.*

## 1. INTRODUCTION

Copper is obtained by pyrometallurgical processing of high-grade sulfide ores, whereby the metal is extracted by smelting and refining. Conventional pyrometallurgical copper extraction processes require high temperatures and emit significant amounts of harmful gasses, making them environmentally and economically unfavorable. Hydrometallurgical processes represent an alternative that enables lower energy costs and less environmental impact. This has led to increased interest in the extraction of copper from low-grade sulfide ores in aqueous solution, where secondary copper sulfides being recognized as an important resource. [1-3]

The most common secondary copper mineral in nature is Covellite (CuS). It is classified as an intermediate product made during the oxidation process of chalcocite ($Cu_2S$) and other complex copper sulfide minerals. Covellite is more stable during oxidation in the presence of sulfur then other copper sulfide minerals. In addition, during the leaching in strongly acidic medium, elementary sulfur may formed passivation layer and block the surface from contact with solution. [4]

Research shows that covellite is leached in different media such as ammonia [5], nitrates [6], chlorides [7-8] and bioleaching [9-10]. Most of the previous studies focused on the dissolution of pure mineral phases or synthetic samples [11-12]. However, in industrial conditions, copper is not obtained from individual minerals, but from ores containing various mineral components, including oxides, sulfides and gangues. Ionic liquids represent a new class of solvents characterized by low vapor pressure, chemical and thermal stability and reusability [13]. Their use in hydrometallurgy has increased significantly in recent years. Numerous studies have shown that ionic liquids can increase the effectiveness of sulfide dissolution, especially in combination with oxidizing agents [14,15]. Dong et al. [14] have shown that Brønsted acid ionic liquids can effectively dissolve chalcopyrite, while Aguirre et al. [15] finds that imidazolium ionic liquids achieve significantly higher extraction in the presence of chloride ions.

In this work, as-received covellite concentrate, obtained from an industrial plant, was used without additional treatment. The results of copper concentrate leaching with the ionic liquid 1-butyl-3-

methyl imidazolium hydrogen sulfate ([bmim]$HSO_4$) in the absence and presence of hydrogen peroxide ($H_2O_2$) are presented.

## 2. EXPERIMENTAL

For the purposes of this research, a sample obtained from the flotation process plant of Serbia Zijin Mining d.o.o. Bor was used. After collection, the sample was dried at room temperature, homogenized, and prepared for further analysis and experiments. The main elements present in the concentrate are copper and iron. Table 1 shows the chemical composition of the sample:

*Table 1. Chemical composition of the copper concentrate*

| Element | Cu | Fe | Zn | Pb | S | $SiO_2$ | CaO | MgO |
|---|---|---|---|---|---|---|---|---|
| % | 15.17 | 25.34 | 0.07 | 0.02 | 36.72 | 8.87 | 0.52 | 0.03 |

**X-ray diffraction analysis was used to determine the phase composition of the samples. The samples were analyzed with a PHILIPS X-ray diffractometer PV-1820/1710 (Cu Kα radiation 0.154178 nm; 40 kV; 30 mA).**

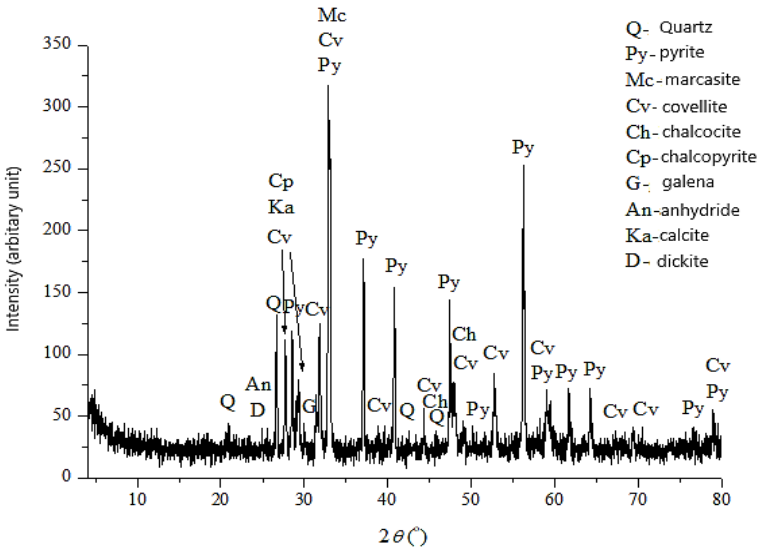


*Figure 1. X-ray diffraction pattern of initial covellite concentrate*

Figure 1 shows that the predominant sulfide minerals are pyrite and covellite. Marcasite, chalcocite, chalcopyrite and galena were also identified in the analyzed sample, but they are present in significantly smaller amounts. The presence of gangue minerals such as quartz, anhydrite, calcite and dickite was also observed.

### 2.1 Experimental procedure

The leaching experiments were carried out in 600 $cm^3$ glass reactor with 100 $cm^3$ leaching solution and 1 g of solid sample. The magnetic stirrer was used and stirring speed was set up to 600 rpm for all experiments. Ionic liquid solution ([bmim]$HSO_4$ concentration of 0.5 mol/$dm^3$ was prepared and used as a leaching reagent. In the experiments where the oxidizing agent was used, hydrogen peroxide was added to the solution at a certain concentration (0.5-3.0 mol/$dm^3$).
After the leaching time of 120 min, 1 $cm^3$ of the solution was taken and diluted to 50 $cm^3$ in a volumetric flask. The concentration of copper in the diluted samples was determined using optical

emission spectrometer with inductively coupled plasma (ICP-OES Optima 8300; Perkin Elmer) and a multiparameter photometer Hanna (HI 83200).

## 3. RESULTS AND DISCUSSION

Table 2 and Figure 2 show the results of the copper concentrate leaching at a concentration of 0.5 mol/ $dm^3$ ionic liquid solution ([bmim]$HSO_4$) without oxidizing agent and in the presence of hydrogen peroxide ($H_2O_2$)concentration 0.5-3.0 mol/$dm^3$.

*Table 2. Effect of different leaching solutions on copper leaching degree from covellite concentrate after 120 minutes of leaching*

| **Leaching conditions** | [bmim]$HSO_4$ concentration (mol/$dm^3$) | $H_2O_2$ concentration (mol/$dm^3$) | Copper leaching degree (%) |
|---|---|---|---|
| [bmim]$HSO_4$ | 0.5 | / | 1.03 |
| [bmim]$HSO_4$ + $H_2O_2$ | 0.5 | 0.5 | 11.70 |
| | | 1.0 | 12.72 |
| | | 2.0 | 20.67 |
| | | 3.0 | 28.97 |

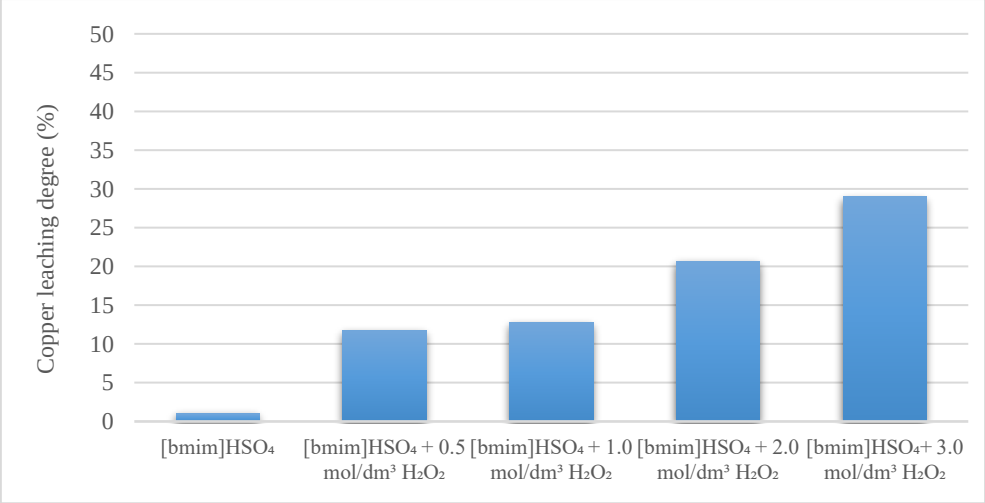


*Figure 2. Influence of $H_2O_2$ concentration on copper extraction from covellite concentrate in ionic liquid solution*

Leaching of the copper concentrate with an ionic liquid solution without hydrogen peroxide was less efficient than leaching in the presence of the oxidising agent. The results indicate that the ionic liquid [bmim]$HSO_4$ solution without oxidising agent is not sufficient for the effective dissolution of covellite, according to the very low copper extraction efficiency (1.03 %) after 120 minutes of reaction time. Previous studies [4] have shown that covellite is difficult to dissolve in acidic media without an oxidising agent, mainly due to the formation of a passive layer of elemental sulfur on the surface of the mineral, which prevents further leaching. The addition of $H_2O_2$ significantly increases the leaching efficiency.

The addition of 0.5 mol/$dm^3$ $H_2O_2$ resulted in an increased copper leaching degree (up to 11.70 %) after 120 minutes of processing time, while only a slight increased occurred at a 1.0 mol/$dm^3$ $H_2O_2$ (12.72 %). With a higher concentration of oxidising agent up to 2.0 mol/$dm^3$ the copper leaching degree significantly enhanced to 20.67%. The copper leaching degree increased to 28.97 % with increasing concentration of the oxidising agent up to 3.0 mol/$dm^3$. These results show that the presence of an oxidising agent plays a crucial role in the extraction of copper ions in solution. The previous studies [3, 7] shows that the dissolution of covellite and other secondary sulfides is limited in the absence of oxidising agents, while the addition of oxidising agents ($H_2O_2$, $Fe^{3+}$, NaClO or $O_2$) significantly accelerates the process.

The results confirm that the ionic liquid solution in combination with hydrogen peroxide can be a promising approach for hydrometallurgical processing of covellite. However, these data were obtained for a single time point (120 min). For a complete kinetic analysis of the process, the dissolution dynamics as a function of time should be investigated.

## 4. CONCLUSION

Leaching experiments were done to dissolve the copper from the covellite concentrate. The results show that the application of the ionic liquid solution ([bmim]$HSO_4$) in the presence of hydrogen peroxide ($H_2O_2$) has a potential for efficient leaching of copper from covellite. The addition of hydrogen peroxide improves the leaching efficiency, with the highest copper leaching degree (28.97%) achieved at 0.5 mol/dm$^3$ [bmim]$HSO_4$ and 3.0 mol/dm$^3$ $H_2O_2$. The copper leaching efficiency in the presence of 0.5 mol/dm$^3$ [bmim]$HSO_4$ without oxidizing agent was 1.03 %. These data indicate the role and importance of the oxidising agent in the oxidation of sulfide minerals.

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