Trans Indian Inst Met
https://doi.org/10.1007/s12666-021-02426-z

ORIGINAL ARTICLE

Dissolution of Chalcopyrite in Presence of Chelating Agent and Hydrogen Peroxide

M. Deniz Turan[1] · J. Pedro Silva[2] · Z. Abidin Sarı[3] · Rashid Nadirov[4] · Norman Toro[5]

Received: 23 February 2021 / Accepted: 16 September 2021
© The Indian Institute of Metals - IIM 2021

Abstract Chelating agents (e.g., EDTA, titriplex III, etc.) are very important salts as they can take a complex form with metal ions. This study investigated usage of titriplex III in presence of hydrogen peroxide for metal extraction from chalcopyrite concentrate under various conditions such as conventional leaching, mechanical activation and ultrasound leaching. The results show that both titriplex and hydrogen peroxide are required for metal extraction from chalcopyrite concentrate. However, mechanical activation is an important parameter for increasing metal extraction, so that copper extraction increases to 83% from 47% after 30 min of grinding chalcopyrite by a high-energy grinding system. Metal extraction efficiency is improved by application of ultrasound to the leaching process. It is possible to obtain copper and iron extraction as 93% and 65%, respectively, under the following leaching conditions: mechanical activation time of 30 min, titriplex concentration of 100 g L^{-1}, H_2O_2 concentration of 2 mol L^{-1}, ultrasound power of 10%, leaching time of 90 min, leaching temperature of 45 °C and liquid–solid ratio of 25 mL g^{-1}.

Keywords Leaching · Titriplex III · Mechanical activation · Chelating · Ultrasound · Hydrogen peroxide · Copper

✉ M. Deniz Turan
mdturan@firat.edu.tr; mdturan@gmail.com

1 Department of Metallurgical and Materials Engineering, Fırat University, 23119 Elazığ, Turkey

2 Institute of Chemistry, UNESP, P.O. Box 355, Araraquara, SP 14800-900, Brazil

3 Department of Metallurgy Iskenderun Vocational School of Higher Education, Iskenderun Technical University, 31200 Iskenderun, Hatay, Turkey

4 Al-Farabi Kazakh National University, 050038 Almaty, Kazakhstan

5 Faculty of Engineering and Architecture, Universidad Arturo Prat, Almirante Juan Jos'e Latorre 2901, 1244260 Antofagasta, Chile

1 Introduction

In general, while sulfide ores are treated by pyrometallurgical methods due to low dissolution and presence of acid-consuming gangue minerals, oxidized ores (carbonates, oxides, sulfates, etc.) are treated by hydrometallurgical methods. One of the most important sources for copper production is chalcopyrite ($CuFeS_2$) that is known as a sulfide ore. Chalcopyrite is crushed and grinded to facilitate flotation, so that liberation of mineral grains requires grinding down to 100 μ. The chalcopyrite concentrate that is obtained from froth flotation is smelted by a high temperature furnace, and the operations that follow may be listed as converting, anode casting and electrowinning [1]. Leaching of chalcopyrite is mostly difficult because of its compact hexagonal crystal structure. In hydrometallurgical copper extraction from chalcopyrite, strong oxidants and acidic leaching conditions are required. Leaching studies of chalcopyrite have been carried out using various leaching agents such as sulfuric acid [2–4], hydrochloric acid and chlorite medium [5, 6], ammonia [7], and various salts [8–10].

In recent years, extraction of metals from various sources has been studied by using new leaching agents. Konishi et al. [11] studied the leaching behavior of chalcocite in an alkaline Na_4EDTA solution containing oxygen. The authors stated that copper leaching was carried out by

Published online: 06 October 2021

chelation of copper (II) with EDTA. It can be said that the addition of organic polar solvents in the aqueous solution causes to higher copper dissolution.

Studies with chelating agents are limited in the literature, but studies using EDTA regarding this topic may be specified as metal leaching from a cathode ray tube [12], thiosulfate leaching of silver sulfide [13] and kinetic-thermodynamic studies of silver leaching [14]. On the other hand, it is observed in literature that chelating agents such as EDTA are mostly used to remove heavy metal pollution [15–17]. Also, as a complementary method, solvometallurgy is an emerging technology [18].

Unfortunately, there is no study on chalcopyrite leaching in presence of titriplex III as a chelating agent. In light of the aforementioned studies, the idea of leaching chalcopyrite using an oxidant and chelating agent with titriplex III is proposed.

This study investigated the leaching behavior of chalcopyrite concentrate with hydrogen peroxide in presence of titriplex III (also known as Na-EDTA) using various leaching methods to increase extraction efficiency at atmospheric conditions.

2 Material and Method

Chalcopyrite concentrate was obtained from Samsun, Turkey. The sample was passed through a 200-mesh sieve, and this fraction was used in all experiments. The sieved sample was dried firstly in the laboratory ambient conditions, then in a furnace at 50 °C for 12 h. The chemical analysis of chalcopyrite concentrate was carried out by the ICP-OES (Inductively Coupled Plasma Optical Emission Spectrometry) method after dissolution by using a microwave oven (Table 1). Mineralogical analysis of the chalcopyrite sample was performed via the powder diffraction technique by an X-ray diffraction (XRD) system. According to the XRD analysis, the concentrate mainly consisted of chalcopyrite ($CuFeS_2$) and pyrite (FeS_2).

The chemicals that were used were 100% titriplex III (ethylenedinitrilotetraacetic acid disodium salt dihydrate-$C_{10}H_{14}N_2O_8$*2Na*2H_2O) and 35% hydrogen peroxide (H_2O_2) without any further purification. Double-distilled water was used in all experiments.

This study examined extraction of metals from a chalcopyrite concentrate with hydrogen peroxide in presence of titriplex III using various leaching methods. For this purpose, the experiments were divided into different groups. The experiments were carried out by (a) leaching with original samples under atmospheric conditions, (b) leaching with mechanically activated samples in a high-energy mill and (c) leaching by using an ultrasound probe.

Atmospheric leaching experiments were performed by using a conventional magnetic multi stirrer (Velp Scientifica MultiStirrer 15). The leaching parameters that were investigated in these experiments are listed as follows: H_2O_2 concentration (0–4 mol L^{-1}), titriplex (III) concentration (10–110 g L^{-1}), leaching temperature (25–65 °C), leaching time (5–420 min) and liquid–solid ratio (5–100 mL g^{-1}).

In order to increase the leaching efficiency, mechanical activation of the chalcopyrite concentrate was achieved in a high-energy ball mill (Model HHS4P-M). The mill contained a tungsten jar with a volume of 80 ml and tungsten balls. In all experiments in this group, the ball to solid ratio was kept constant at 10. 8 g of chalcopyrite concentrate and approximate 80 g ball was fed into milling vessel. After milling process was finished, balls were removed from the milled material and it was then used for leaching experiments.

Ultrasonic leaching experiments were performed with an ultrasound probe that was immersed into the solution. The experiments utilized an ultrasonic generator (Bandelin UW-2070) equipped with a stainless-steel probe, glass beaker, temperature circulator and reflux condenser. The effects of leaching time (5–120 min) and leaching temperature (25–65 °C) in the presence of a constant 10% ultrasound power were investigated.

The resulting leaching solutions were diluted with distilled water and analyzed by the AAS (Atomic Absorption Spectroscopy) method (PerkinElmer, AAnalyst 400).

3 Results

3.1 Theoretical Approach

Leaching of chalcopyrite requires strong oxidants and effective leaching conditions because it is insoluble compound, and it has strong chemical bonds difficult to break. To increase the dissolution rate, researchers have focused on some combined methods. These methods include mechanical activation and pressure leaching [8, 19–21].

Table 1 Chemical analysis of chalcopyrite concentrate

Compound	SiO_2	Cu	Fe	S	Zn	Pb
w/%	1.31	20.50	34.97	35.88	5.09	0.86

Chelating agents have multiple complexation sites in their molecule structure able to interact with metal cations, often yielding stable compounds soluble in water. Moreover, usage of chelating chemicals as leaching agents may be worth examining because the chelating effects may cause a high dissolution rate.

The most important characteristic of titriplex III that has been used in this study is that it can be take a complex form with many metal ions. $CuFeS_2$ leaching processes using H_2O_2 and titriplex III can be summarized by following reactions;

– Generation of O_2 by H_2O_2 decomposition;

$$4H_2O_2(aq) \rightarrow 2\ H_2O\ (l) + O_2(g) \quad (1)$$

– Oxidation of chalcopyrite by O_2;

$$CuFeS_2(s) + O_2 + 4\ H^+ \rightarrow 2\ H_2O(l) + 2S(s) + Cu^{2+}(aq) + Fe^{2+}(aq) \quad (2)$$

– Chelating copper with titriplex III;

$$Cu^{2+}(aq) + Na_2EDTA \rightarrow CuEDTA + 2Na^+(aq) \quad (3)$$

– Leaching reaction of chalcopyrite in presence of titriplex (III) and hydrogen peroxide;

$$CuFeS_2 + 2.5\ H_2O_2 + 2Na_2(EDTA)^{2-} \rightarrow Cu(EDTA)^{2-} + Fe(EDTA)^{2-} + 2S(s)\ 5H_2O + 4Na^+_{(aq)} \quad (4)$$

It is estimated that the presence of titriplex III in the leaching solution helps to decrease the H_2O_2 decomposition due to the formation of highly stable copper–EDTA and iron–EDTA complexes. On the other hand, suggesting that EDTA inhibits the catalytic role of cupric ion through the formation of the Cu complex $(EDTA)^{2-}$; this behavior is unlike that of iron, according to Koppenol and Butler [22] where the Fe (II, III)–EDTA complexes formed in the leach liquor can react with the hydrogen peroxide (Eqs. 5 and 6).

$$Fe(EDTA)^- + 2H_2O_2 \rightarrow Fe(EDTA)^{2-} + OH^* + O_2 + H_2O + H^+ \quad (5)$$

$$Fe(EDTA)^{2-} + H_2O_2 + H^+ \rightarrow Fe(EDTA)^- + OH^* + H_2O \quad (6)$$

Thermodynamic stability has an important role in determining the bond strength between metal ligands. Some complexes are stable, but as soon as they are introduced into aqueous solution, it is seen that these complexes have an effect on stability and they fall apart. The values of stability constant increases with a decrease in the concentration of ligands and shows a high stability constant of metal–ligand complex at a very low concentration of ligands. The high stability constant may be attributed to high complexation between metal ion and ligand. The stability constant values for copper and iron complexes shows the trend 14.3°18.8°25.7. The Fe(III)–EDTA complex is more stable than the Fe(II)–EDTA complex. However, the Cu(II)–EDTA complex is more stable than the Fe(II)–EDTA complex. This case indicates that copper and iron(III) tend to have a higher complexation in this study.

On the other hand, thermodynamic stability constants for copper and iron complexes with EDTA are given in Table 2. However, the leaching reaction of chalcopyrite contains complex processes involving multiple steps converting copper species into bornite, covellite or chalcocite based on leaching conditions. Furthermore, cationic exchange may be possible between titriplex III and Fe ions that are extracted from chalcopyrite.

3.2 Experimental

3.2.1 Leaching Behavior of the Original Sample

The effects of titriplex III concentration were investigated in the range of 10–110 g L^{-1}. As shown in Fig. 1, copper extraction first increased with increasing titriplex III concentration, while afterward, it decreased despite the increasing chelating agent concentration. It is clear that the compact crystal structure of chalcopyrite could not be degraded under this leaching condition.

Figure 2 shows the effects of hydrogen peroxide concentration on the metal extraction process. As seen in Fig. 2, copper extraction noticeably increases with increasing hydrogen peroxide concentration, while iron dissolution significantly decreases with increasing hydrogen peroxide after the concentration of 2 mol L^{-1} of H_2O_2. This trend may be explained by the formation of low-solubility iron oxide compounds with the increased hydrogen peroxide amount that supply more oxygen to the leach solution. This figure also indicates that it is possible to obtain selective leaching by keeping hydrogen peroxide concentrations high.

Nevertheless, hydrogen peroxide concentration has been chosen as 2 mol L^{-1}. The maximum iron dissolute level at this concentration will allow determining the leaching behavior of the dominant metals (Cu and Fe) in presence of the chelating reagent.

According to the results of the experiment so far, it is seen that titriplex III and hydrogen peroxide presence is required. It is clear that metal dissolution is quite limited without both reactants in the leaching solution.

The effects of leaching temperature are shown in Fig. 3. It is observed that metal extraction first increases with

Table 2 Thermodynamic stability constants for copper and iron complexes with EDTA

Complex	Stability constants, log *K*
Cu (II)-EDTA	18.8
Fe (II)-EDTA	14.3
Fe (III)-EDTA	25.7

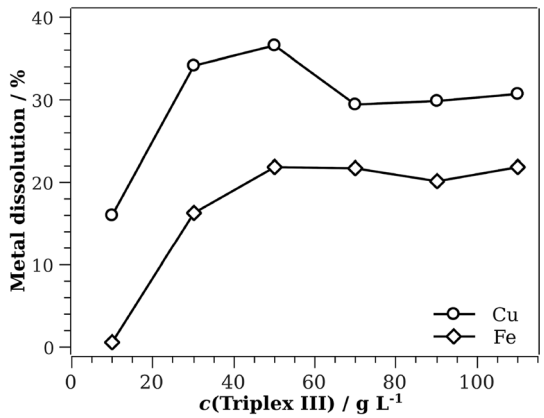

Fig. 1 Effect of titriplex III concentration on the metal extraction using H_2O_2 1 mol L^{-1}, 45 °C of leaching temperature, 400 rpm of stirring rate, 120 min of leaching time and 25 mL g^{-1} of liquid–solid ratio

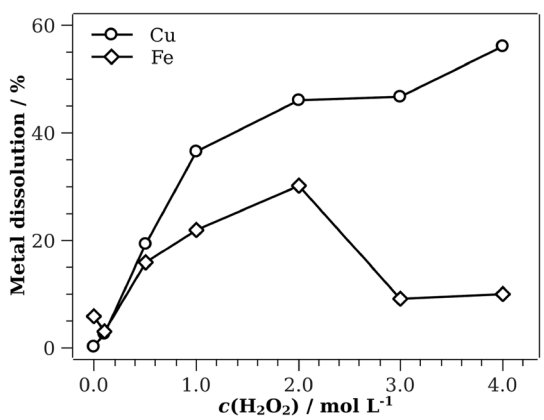

Fig. 2 Effect of hydrogen peroxide concentration on the metal extraction, using 50 g L^{-1} of titriplex III concentration, 45 °C of leaching temperature, 400 rpm of stirring rate, 120 min of leaching time and, 25 mL g^{-1} of liquid–solid ratio

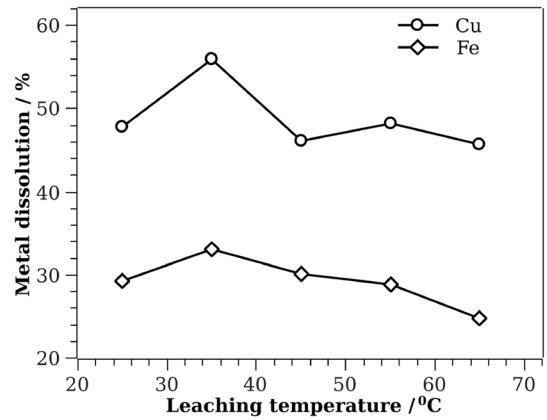

Fig. 3 Effect of leaching temperature on the metal extraction using 50 g L^{-1} of titriplex III concentration, 2 mol L^{-1} of hydrogen peroxide, 400 rpm of stirring rate, 120 min of leaching time and 25 mL g^{-1} of liquid–solid ratio

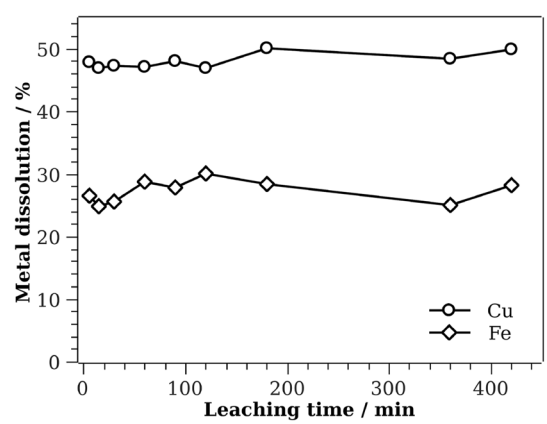

Fig. 4 Effect of leaching time on the metal extraction, using 50 g L^{-1} of titriplex III concentration, 2 mol L^{-1} of hydrogen peroxide, 400 rpm of stirring rate, 45 °C of leaching temperature and 25 mL g^{-1} of liquid–solid ratio

increasing the leaching temperature, and then, it starts to decrease. This situation may be explained by the increasing decomposition rate of H_2O_2 with temperature. Active oxygen released from the rapid decomposition of H_2O_2 under the atmospheric conditions may escape from the leaching medium without achieving further oxidation.

Figure 4 shows the effects of leaching time on metal extraction. The results that are obtained indicate that leaching time is not a parameter that has influence on the metal extraction process, such that the leaching reaction achieve in a short time an almost static dissolution condition around 50% to Cu and 30% to Fe.

Figure 5 presents the effects of liquid–solid ratio on metal extraction. As expected, metal extraction increases with the increase in the liquid–solid ratio. In these experiments, the reason for being able to achieve high metal extraction with the increasing liquid–solid ratio is that, the solution volume is kept constant, and the amount of solids

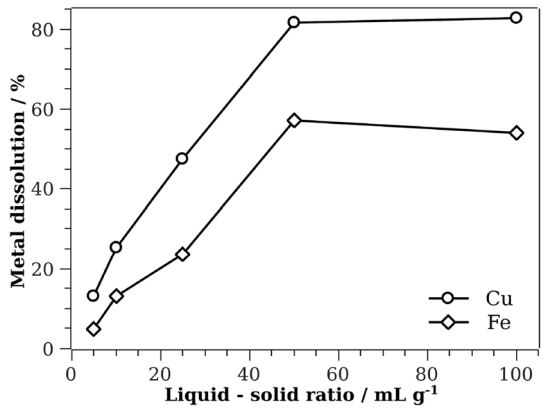

Fig. 5 Effect of liquid–solid ratio on the metal extraction, using 50 g L^{-1} of titriplex III concentration, 2 mol L^{-1} of hydrogen peroxide, 400 rpm of stirring rate, 45 °C of leaching temperature and 30 min of leaching time

is changed. Thus, the amount of solution per unit solid increases, although lower metal concentration is obtained in the solution.

The results of the experiments demonstrate that limited copper (47%) and iron (24%) extraction is obtained under the following leaching conditions: 50 g L^{-1} of titriplex III, 2 mol L^{-1} of H_2O_2, 45 °C of leaching temperature, 30 min of leaching time and 25 mL g^{-1} of liquid–solid ratio.

In the next part of the study, the effects of mechanical activation and ultrasound leaching of the mechanically activated samples on the metal extraction process are described.

The purpose of these experiments is not only to increase extraction efficiency but also to determine the leaching behavior of the chelating agent on (1) chalcopyrite with an activated particle surface, (2) chalcopyrite with a cavitated particle surface by ultrasound.

3.2.2 Leaching Behavior of Treatment Sample

Mechanical activation method maintains the mechanical energy of material surface. Thus, new activated surfaces can be obtained on particle surface so that this material can easily reacts with chemical reagents. The resulting creation of fine particles, the increase in specific surface area and the formation of defective structures accelerate leaching in hydrometallurgy [23].

The chalcopyrite concentrate is grinded in a high-energy mill system at different times and then leached under certain conditions. However, 100 g L^{-1} of titriplex III concentration has been used for leaching the mechanically activated samples. This is because, the activated particle surface catalyzes the decomposition of hydrogen peroxide in an uncontrolled manner under the condition of using 50 g L^{-1} of titriplex III, so that the use of greater amounts of titriplex III (known to have a long molecular chain) in the solution assumes the role of a stabilizer for hydrogen peroxide. On the other hand, it is observed that metal extraction decreases after 30 min of grinding time (Fig. 6). This situation may be attributed to agglomerate formation, which causes a decrease in particle surface area [24]. The extraction rates of copper and iron under the optimum leaching conditions (30 min of grinding time and 100 g L^{-1} of titriplex III) have been calculated as 83% and 58%, respectively.

If mechanical activation causes a chemical transformation in the material, this is called a mechanochemical activation. The diffraction graphs of the samples with original concentration and the one mechanically activated for 30 min is given respectively in Figs. 7 and 8 for evaluation according to X-ray peaks. Figure 8 XRD results indicate the presence of chalcopyrite ($CuFeS_2$), chalcocite (Cu_2S) and cubanite ($CuFeS_3$). Moreover, it seems that one of the main mineral phases (pyrite-FeS_2) maintain its mineralogical form after mechanical activation. The grinding process applied on chalcopyrite also lead to an increased reaction yield by causing it to react readily with the leaching reactants.

The SEM images of the mechanically activated sample at different magnification rates are shown in Fig. 9. The images include the mechanical activation traces of chalcopyrite related to how the structural defects in particles occur. Furthermore, the circular markings on the image indicate the initiation of agglomeration on the particle surface.

In leaching experiments with ultrasound (US) waves, the cavitation effect on the solid particle surface has been discussed [25]. This cavitation effect attributes; breaking up large ore particles to increase the particle diffusion and mass transfer rate, and also remove the formed elemental

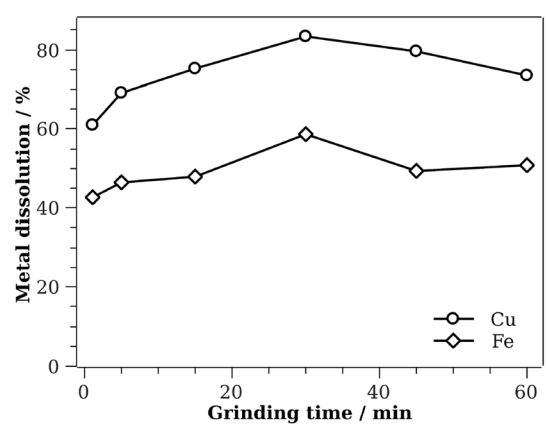

Fig. 6 Effect of mechanical activation time, using 100 g L^{-1} of titriplex III concentration, 2 mol L^{-1} of hydrogen peroxide, 400 rpm of stirring rate, 45 °C of leaching temperature and 30 min of leaching time

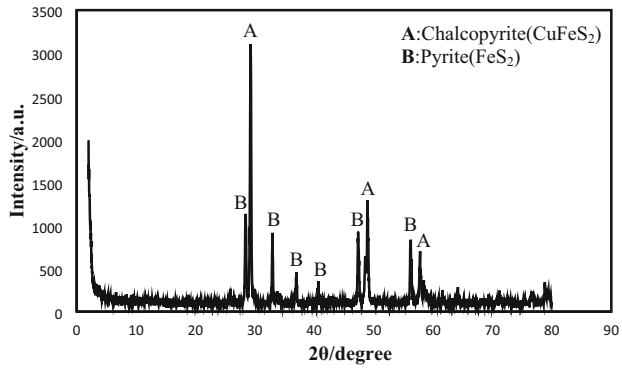

Fig. 7 XRD pattern of original chalcopyrite concentrate

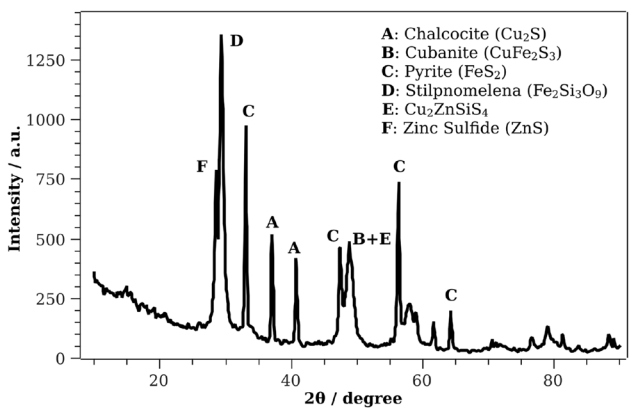

Fig. 8 XRD pattern of mechanically activated chalcopyrite concentrate after 30 min of grinding

sulfur layer to enhance the chemical reactions for more sulfide minerals dissolution. Advanced oxidation processes (AOPs) such as US, US/H_2O_2, produce oxidizing species, generally hydroxyl radicals which are very powerful and unselectively oxidize inorganic sulfide minerals compounds. The rapid decomposition of hydrogen peroxide may also observe the presence of US in aqueous solution. Here, US plays an essential role in the formation of reactive species (H_2O_2, $OH^{\cdot}$, $H^{\cdot}$, $O^{\cdot}$ and $HO_2^{\cdot}$).

A probe that creates sound waves in a leaching solution creates a good mixing effect. When ultrasonic energy is emitted by a solid, its effects on the solid surface depend on the conditions in the medium.

Ultrasound power is kept constant at 10% in all experiments to prevent rapid decomposition of hydrogen peroxide. The effects of leaching temperature in presence of ultrasound on the mechanically activated material may be seen in Fig. 10. This figure demonstrates that the metal extraction rate does not change with increased leaching temperature. As an interesting finding, it is seen that metal extraction does not decrease with increasing leaching temperature in the presence of ultrasound, in contrast to the case in magnetic stirring. In this case, even in high

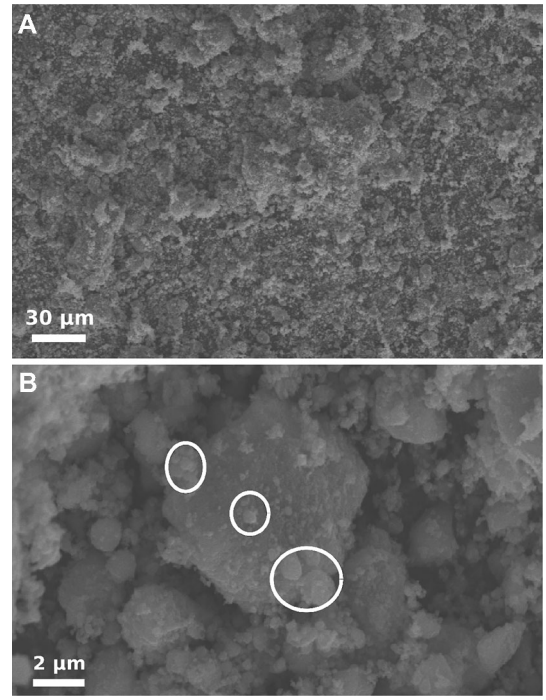

Fig. 9 SEM images of mechanically activated chalcopyrite concentrate grinded for 30 min, obtained at magnification of 100x (**a**) and 15000x (**b**)

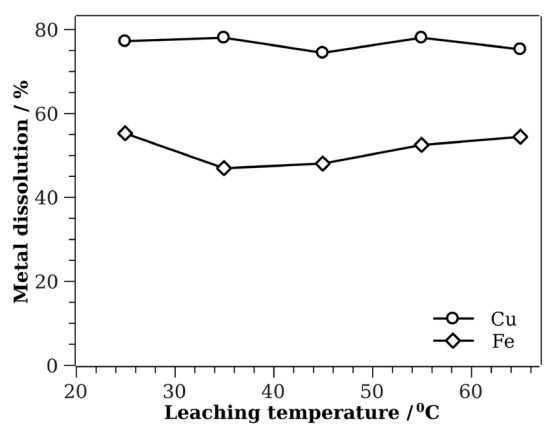

Fig. 10 Effect of leaching temperature by treatment of ultrasound using mechanical activation for 30 min, ultrasound power: 10%, leaching time of 30 min, titriplex III concentration of 100 g L^{-1} and H_2O_2 concentration of 2 mol L^{-1}

temperature experiments, H_2O_2 is decomposed in a controlled manner in the presence of US during the reaction. Also, an increase in leaching efficiency could be attributed to ultrasound treatment decreasing the resistance to mass transfer in the system or removing insoluble substances from the solid surfaces. On the other hand, high degree of structural disorder, the mechanical activation of minerals makes it possible to reduce their decomposition temperature.

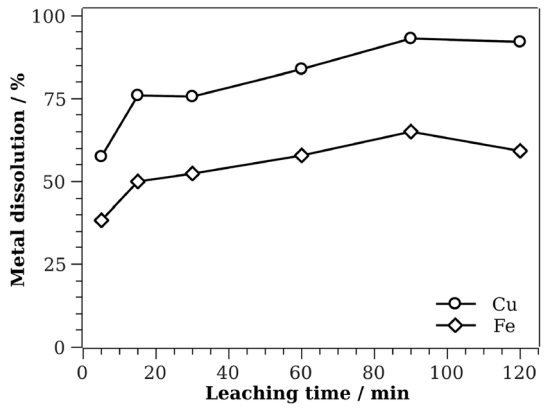

Fig. 11 Effect of leaching time by treatment of ultrasound using 30 min of mechanical activation, ultrasound power of 10%, leaching temperature of 45 °C, 100 g L^{-1} of titriplex III and H_2O_2 2 mol L^{-1}

Figure 11 shows that metal extraction increases with increasing leaching time until 90 min, then reaches a limit. It is obvious that copper extraction exceedes 90% from the mechanically activated chalcopyrite concentrate with hydrogen peroxide and titriplex III in presence of ultrasound.

The results of the SEM analysis on the leach residue are shown in Fig. 12. The SEM images indicate that the amount of large particles decreases as a result of leaching conditions.

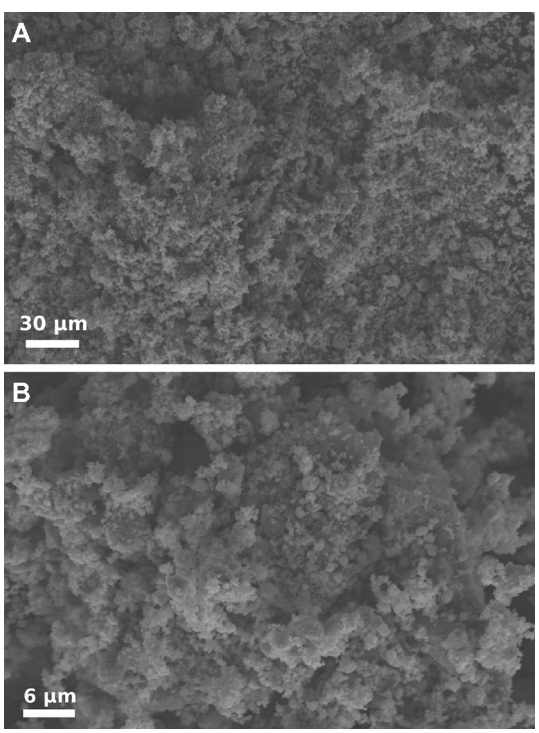

Fig. 12 SEM images of leach residue, submitted to the following experimental conditions: mechanical activation time of 30 min, ultrasound power of 10%, leaching temperature of 45 °C, 100 g L^{-1} of titriplex III and H_2O_2 2 mol L^{-1}, obtained at magnifications of 100x (**a**) and 500x (**b**)

4 Conclusion

The effects of hydrogen peroxide and titriplex (III) on metal extraction from chalcopyrite concentrate were investigated. The effects of the chelating agent in presence of hydrogen peroxide were investigated with conventional leaching conditions, mechanically activated material and ultrasound conditions. According to the results that were obtained, presence of both hydrogen peroxide and titriplex III was required for leaching. Hydrogen peroxide rapidly decomposed, and isothermal leaching conditions disappeared in the presence of copper and iron. In order to overcome this problem, the use of titriplex (III) was provided in all experiments as a reactant to slow down the degradation rate of hydrogen peroxide and form complexes with soluble metal ions. It was determined that metal extraction increases with increased titriplex III and hydrogen peroxide concentrations. However, iron extraction markedly decreased from about 30% to 9% at concentrations beyond 2 mol L^{-1} of H_2O_2. As a result of the use of ultrasound waves in the leaching experiments, it was understood that the same metal extraction rate was achieved at lower temperatures than the traditional leaching conditions. The highest metal extraction (around 93% Cu and 65% Fe) rates were obtained under the following conditions: mechanical activation time: 30 min, leaching time: 90 min, ultrasound power: 10%, leaching temperature: 45 °C, titriplex III concentration: 100 g L^{-1}, H_2O_2 concentration: 2 mol L^{-1} and liquid–solid ratio: 25 mL g^{-1}.

Acknowledgements The author wishes to express thanks to metallurgical and material engineers Ahmet Burak Deveden and Selim Coşkunırmak for their help in conducting the experiments.

References

1. Davenport W G, King M, Schlesinger M, and Biswas A K, *Extractive Metallurgy of Copper*, Elsevier Science, Oxford (2002).
2. Dreisinger D, and Abed N, *Hydrometallurgy* **66** (2002) 37.
3. Han K N, and Meng X, *Min Metal Proces* **20** (2003)160.
4. Turan M D, and Altundoğan H S, *Met Mat Trans B* **44B** (2013) 809.
5. Cai Y F, Chen X M, Ding J Y, and Zhou D S, *Hydrometallurgy* **113–114** (2012) 109.
6. Liddicoat J, and Dreisinger D, *Hydrometallurgy* **89** (2007) 323.
7. Feng D, and Van Deventer J S J, *Hydrometallurgy* **63** (2002) 189.
8. Turan M D, and Altundoğan H S, *Int J Min Met Mat* **21** (2014) 862.
9. Turan M D, *Canadian Met Quart* **53** (2014) 444.
10. Turan M D, Boyrazlı M, and Altundoğan H S, *J Cent South Univ* **25** (2018) 21.

11. Konishi Y, Katoh M, and Asai S, *Metal Trans B* **22B** (1991) 295.
12. Pant D, Singh P, and Upreti M K, *Hydrometallurgy* **146** (2014) 89.
13. Siller D M P, Aceituno J C F, and Alonso F N, *Hydrometallurgy* **149** (2014) 1.
14. Siller D M P, Aceituno J C F, and Alonso F N, *Hydrometallurgy* **134–135** (2013) 124.
15. Wasay S A, Barrington S F, and Tokunaga S, *Env Tech* **19** (1998) 369.
16. Pociecha M, and Lestan D, *J Hazar Mat B* **165** (2009) 533.
17. Palma L D, and Ferrantelli P, *J Hazar Mat B* **122** (2005) 85.
18. Li X, Monnens W, Li Z, Fransaer J, and Binnemans K, *Green Chem* **22** (2020) 417.
19. Tkacova K, and Balaz P, *Int J Min Proc* **44–45** (1996) 197.
20. Wodka J, Chmielewski T, and Ziolkowski B, *Phys Prob Min Proc* **41** (2007) 349.
21. Turan M D, Arslanoğlu H, and Altundoğan H S, *J Taiwan Ins Chem Eng* **50** (2015) 49.
22. Koppenol W H, and Butler J, *Adv Free Radic Biol Med* **1** (1985) 91.
23. Turan M D, and Balaz P, *J Mining Sci* **56** (2020) 288.
24. Baláž P, *Extractive Metallurgy of Activated Minerals*. Elsevier Science B.V, Amsterdam (2000)
25. Borkent B M, Arora M, and Ohl C D, *J Acous Soc Am* **121** (2007) 1406.

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.