

A Comparison of Galvanic Interactions Between $\text{CuFeS}_2/\text{FeS}_2$ and $\text{CuFeS}_2/\text{PbS}$ During the Kinetic Dissolution of Cu

Reitumetse Tutubalang and Elvis Fosso-Kankeu

Abstract— Leaching copper from chalcopyrite (CuFeS_2) has proven difficult because of surface passivation and poor dissolving rates. With an emphasis on leaching conditions, redox potential, and temperature impacts, this study examines the effects of galvanic interactions between chalcopyrite and galena (PbS / PbS) and chalcopyrite and pyrite (FeS_2 / FeS_2) on copper dissolving rates. Chalcopyrite leached alone and with pyrite or galena at 4:1 mass ratio was used in the experiments. The leaching medium was sulphuric acid, and the temperatures were 25°C, 35°C, 45°C, and 55°C. The redox potentials were approximately 310–320 mV. The findings showed that temperature had a favourable impact on dissolving rates; for chalcopyrite alone, the maximum Cu recovery was 67% at 55°C. The inclusion of galena and pyrite, however, unexpectedly reduced the recovery of Cu at 55°C, the greatest recovery with pyrite was 38%, but the maximum recovery with galena was 13%. Chalcopyrite alone exhibited the highest activation energy (E_a) of 19.69 kJ/mol, according to the research. When combined with pyrite and galena, the E_a dropped to -5.26 kJ/mol and -24.90 kJ/mol, respectively, indicating a change to diffusion control. In conclusion, galvanic interactions with galena or pyrite did not improve copper recovery as anticipated. This is probably because there were no oxidants and insufficient redox potential, which restricted electron transport and made passivation easier. To enhance copper recovery in such systems, future research should investigate introducing oxidants such as ferric ions and optimising redox potential.

Keywords— Chalcopyrite, Hydrometallurgy, Leaching, Galvanic Couples, Shrinking Core Model, Rate Limiting Factor.

I. INTRODUCTION

Since from the past century copper has been one of the most widely used metals worldwide, with a range of applications across various industries such as electric wiring, production of coins, piping & etc. Chalcopyrite (CuFeS_2) is one of the most prevalent and economically important minerals of copper among many other sulphides. The dissolution of copper (Cu) from chalcopyrite, a mineral composed of copper, iron, and sulphur (CuFeS_2), is a process of significant interest in metallurgy and extractive industries. While chalcopyrite is the main supply of copper in the world, it has been important to understand how it dissolves to create

efficient techniques for extracting copper [1-8]. Although chalcopyrite has been identified as major source of copper, it is difficult to extract copper from it because it is easily passivated, resulting in low dissolution kinetics and copper extraction. Industrial applications of chalcopyrite leaching develop slowly because of the low dissolution rate, which is mainly caused by the passivation layer covered on chalcopyrite surface. Passivation is a chemical process that protects materials, usually metals, from corrosion by creating a passive or less reactive surface layer. This layer acts as a barrier, hindering the leaching solution from reaching the underlying copper, slowing down the dissolution rate and recovering minimum copper [9].

The reported possible passivating species are mainly consisted of polysulfide (S^{2-}), elemental sulphur (S) and insoluble sulphate (SO_4^{2-}). It was found that metal-deficient polysulfide was the main passivation substance, but Ag^+ or silver containing species could be effective in increasing the conductivity of polysulfide and then eliminate the passivation effect. Sulphur-oxidizing microorganisms are utilised in the hydrometallurgical treatment of the CuFeS_2 mineral as a strategy to overcome passivation and enhance metal recovery by promoting the oxidation of elemental sulphur [10]. Nevertheless, galvanic interactions between CuFeS_2 and other minerals are the focus of the second technique. Galvanic interactions refer to the electrochemical phenomena that occur when two or more dissimilar metals or minerals come into electrical contact in the presence of an electrolyte [11]. Because of the dissimilar electrochemical properties of the minerals, this contact generates a potential difference that causes electrons to flow from one mineral (the anode) to the other (the cathode) which promotes accelerated dissolution and protection, respectively. A mineral oxidises more quickly in a galvanic couple when it serves as the anode than it would in isolation [12]. Increased corrosion or dissolving results from this faster oxidation. The result of galvanic interaction is to change the rates of the anodic and cathodic half-reactions that occur at the surface of each mineral.

Minimization of passivation during dissolution of copper from chalcopyrite is of important in this study. Consideration of minimizing the impact of passivation during the dissolution of copper from chalcopyrite would not only help with increasing the dissolution rate but also with maximizing the recovery of

Reitumetse Tutubalang¹, Department of Metallurgy at the University of Johannesburg in South Africa.

Elvis Fosso-Kankeu², Department of Metallurgy at the University of Johannesburg in South Africa

copper. Since chalcopyrite is a semiconductor, and its surface reactions during dissolution in aqueous solutions are electrochemical in nature, these enables the investigation and operation of chalcopyrite dissolution using electrochemical approaches.

II. METHODOLOGY

A. Preparation of solutions

Solutions of the pH of 1.5 were prepared using analytical reagent-grade sulphuric acid (H_2SO_4 98% A.C.E.), and deionized water. The redox potential (Ag/AgCl) measurements were determined using a platinum electrode in reference to the Standard Hydrogen Electrode (SHE).

B. Chalcopyrite, Pyrite and Galena Sample Characterization

The $CuFeS_2$ sample obtained as a concentrate from the Phalaborwa Copper Mining Company was used. The pyrite and galena were also obtained as concentrates. All these three samples were dried out moisture using an oven at $100^\circ C$ and later were sub-sampled. For a dissolution feed, grains smaller than 75 microns were used. Chalcopyrite samples were taken for analyses. For determining elemental composition of chalcopyrite X-ray Fluorescence (XRF) (Rigaku-ZSX Primus II with SQX analysis software (Japan)) equipment was used. The elemental composition (XRF) powder method was carried out on a Rigaku-ZSX Primus II in conjunction with the S.Q.X. analysis software (Rigaku, Japan), operating at 4 kW, 60 kV and 150 mA. The solutions collected during the leaching process were analysed for the detection of copper using Atomic Absorption Spectroscopy (AAS).

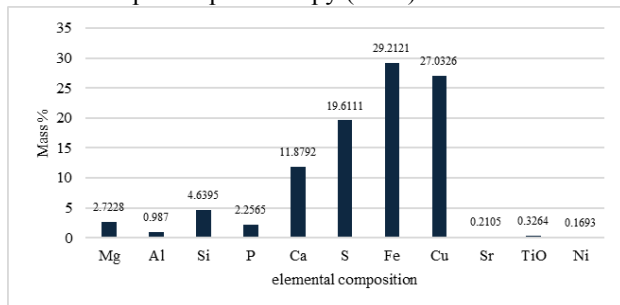


Fig. 1: XRF composition for chalcopyrite sample

XRF analysis was conducted to determine chemical analysis of chalcopyrite. Chalcopyrite sample contained 27.032% copper, 29.212% iron, 19.611% sulphur, 11.879% calcium and 4.639% of silicon. Ni, Sr, Al, and P were found to be trace elements.

C. Leaching process

Leaching process was in a 400ml beaker with a solution of 100mL for each compartment. A ratio of 4:1 for both $FeS_2/CuFeS_2$ and Pb/S was used to evaluate the effect of FeS_2 or PbS addition on Cu dissolution from chalcopyrite. A pulp was made with deionised water and solids. 4g of $CuFeS_2$ was dissolved in acidified sulphuric acid (H_2SO_4) with and without

FeS_2 and PbS addition, separately. Homogenous mixing of the pulp was achieved by agitation with a propeller stirrer at a rotation speed of 350 rpm. The media pH was measured and maintained at 1.5 by adding sodium hydroxide. Initial redox potential was measured and was approximately 310mV. The process was performed at different range of temperatures being $25, 35^\circ C, 45^\circ C$ and $55^\circ C$. The period of the leaching process was 4 hours. A pulp of 10% solids was used and 10ml of solution was withdrawn from a beaker every after 30min.

After leaching process, the solution was filtered using a filter paper to remove the solids and the solution was analysed using AAS for chemical analysis of Cu in the solution. Thereafter, the results were interpreted.

III. RESULTS AND DISCUSSION

A. Cu dissolution curves and recovery

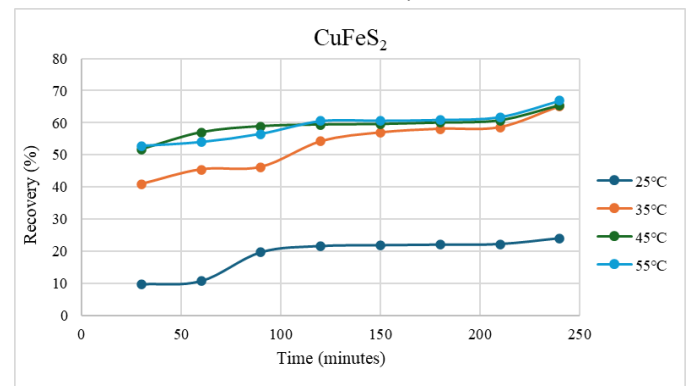


Fig. 2: Recovery vs time graph for $CuFeS_2$ only

Copper dissolution data were obtained from leaching mixtures of 4:1 proportion of the pyrite concentrate and chalcopyrite concentrate. Figure 1 displays the findings. It should be noted that the fraction of copper in solution nearly matched the chalcopyrite dissolution since chalcopyrite was the primary mineral in the solids that contained copper. Figure 1 shows the Cu dissolution % depended on temperature when it was leached alone without addition of pyrite or galena. It is evident that when temperatures were increased in increment of $10^\circ C$, the Cu dissolution % also increased. The highest achieved Cu dissolution was approximately 67% at $55^\circ C$ after 4 hours and the lowest was 10% at $25^\circ C$ with a gradual increase over time. At $25^\circ C$ the recovery of Cu was the slowest, however at $35^\circ C$ the recovery rate is noticeably faster than at $25^\circ C$, reaching the plateau around 200 minutes. Lastly, the recovery rate is even faster at $45^\circ C$, with the plateau reached earlier within 90 minutes.

B. Effect of pyrite on Cu dissolution

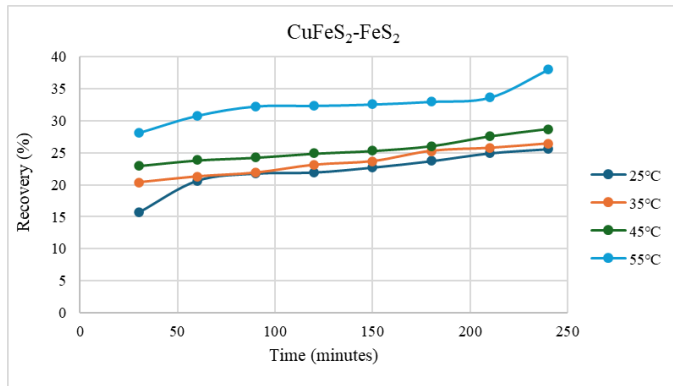


Fig. 4: Recovery vs time graph for CuFeS₂-FeS₂

During the second round of leaching, chalcopyrite was mixed with pyrite (FeS₂-CuFeS₂) at a mass ratio of 4:1. In figure 2, it was observed that the recovery percentage increases with time for all temperatures used during leaching process. The Cu dissolution at 25°C, 35°C and 45°C was found to not have a huge % difference from one another. However, the highest Cu % recovery achieved when CuFeS₂ was mixed with FeS₂ it was found to be 38% at 55°C which is approximately half of the highest Cu % recovery of CuFeS₂ when it was leached without the addition of FeS₂. At 45°C the highest Cu % recovery was approximately 29% and the lowest being at 23%. At 35°C and 25°C, the highest Cu % recovery was found to be nearly 27% and the lowest were 21% and 16%, respectively. The results from the present investigation show that the FeS₂ addition doesn't positively affect the dissolution of CuFeS₂ rate and recovery because the Cu dissolved when CuFeS₂ was coupled with FeS₂ was less compared to when CuFeS₂ was leached without the addition of FeS₂.

C. Effect of Galena on Cu dissolution

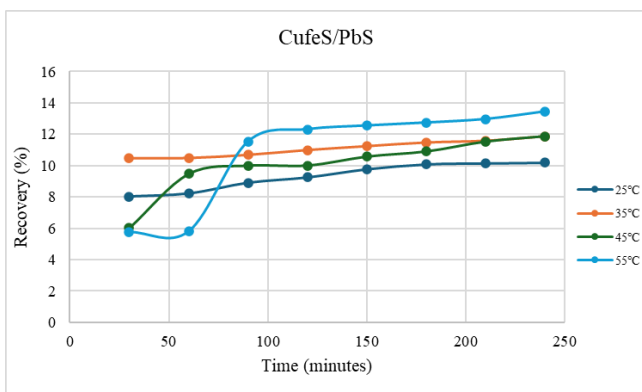


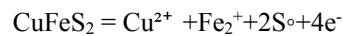
Fig. 4: Recovery vs time graph for CuFeS₂-PbS

During the second round of leaching, chalcopyrite was mixed with pyrite (PbS-CuFeS₂) at a mass ratio of 4:1. In figure 2, it was observed that the recovery percentage increases with time for all temperatures used during leaching process. From figure 3, it is evident that the Cu % recovery achieved was low. At 55°C the highest Cu % recovery was

found to be approximately 13%, the lowest being 6%. For 45°C and 35°C, the highest Cu % recovery obtained was 12%, with the lowest being approximately 6% and 10%, respectively. Lastly at 25°C, the highest Cu dissolution was approximately 10% with the lowest being 8%. It is worth highlighting that the results showed that PbS addition affect negatively the dissolution of CuFeS₂ rate and recovery because when leaching mixture of PbS- CuFeS₂, lowest % of dissolved Cu were obtained.

According to literature, pyrite addition has a major catalytic effect on the chalcopyrite leaching due to the formation of a galvanic cell between the minerals which is caused by their difference in rest potential. However, the results from the experiment conducted does not agree with what literature says. There might be possible factors that might have contributed to pyrite to not have a major catalytic effect on the chalcopyrite leaching which resulted to obtaining less % Cu recovery. Some of the factors might be due to lack of significant galvanic interaction, redox potential, lack of oxidant like ferric irons and passive layer on the surface of chalcopyrite, Ph and local acidification. Due to minerals having different equilibrium potentials CuFeS₂-0,56V and FeS₂-0,66V, galvanic interactions between these minerals can occur due to their different electrochemical potentials [13]. This interaction affects the dissolution behaviour of chalcopyrite, typically resulting in either an increased or decreased rate of copper dissolution, depending on specific conditions.

According to Liu et al [14], in the galvanic couple between CuFeS₂- FeS₂, chalcopyrite undergoes oxidation leading to the release of Cu²⁺ ions into solution. Chalcopyrite dissolution in acidic media is an electrochemical reaction, therefore it can be written as anodic half reaction:



At the cathode, the electrons released from chalcopyrite are accepted by pyrite. During the experiment because of ferric irons (Fe³⁺) were not used as oxidant, proton (H⁺) from sulphuric acid might have been produced. During the process of leaching CuFeS₂ coupled with FeS₂, there must be controlled conditions for galvanic interactions to be significant. According to Olvera et al [15] the optimal oxidising potential for chalcopyrite ranges from 400-470 Mv, however the redox potential that was used during the experiment was around 310-320 Mv. The gap between the optimal redox potential and the one that was used during the experiment might be one of the reasons why the Cu dissolution was low when CuFeS₂ coupled with FeS₂, because the oxidizing environment at this Eh value doesn't allow galvanic coupling between pyrite and chalcopyrite, where pyrite acts as the cathode (accepting electrons), enhancing the dissolution of chalcopyrite at the anode. Therefore, there were less interactions that lowers the energy barrier for chalcopyrite oxidation, effectively boosting the copper recovery rate.

In addition to the Eh used being low than the optimal, there were no oxidising agents that were used during the leaching process. Comparing the % Cu recovery results obtained from

the experiment and the ones from Koleinis (2017) experiment, it was evident that the initial concentration of ferric irons used as an oxidant plays a huge role in recovery of Cu^{2+} because they are primary oxidant therefore, they initiate the oxidation of chalcopyrite, which is typically stable and difficult to dissolve. Consequently, the absence of ferric irons during the leaching process of CuFeS_2 coupled with FeS_2 might be another reason why the % Cu recovery was low as compared to the one obtained in Koleinis [16] experiment.

The reason for a high recovery of 67% when CuFeS_2 was leached without the addition of FeS_2 might be because of when chalcopyrite was reacting alone there were enough H^+ ions from sulphuric acid to oxidise it. Since there were less significant galvanic interactions between CuFeS_2 - FeS_2 , there are possibilities that both pyrite and chalcopyrite were undergoing oxidation therefore there was a competition for H^+ ions thus less Cu dissolution in the solution.

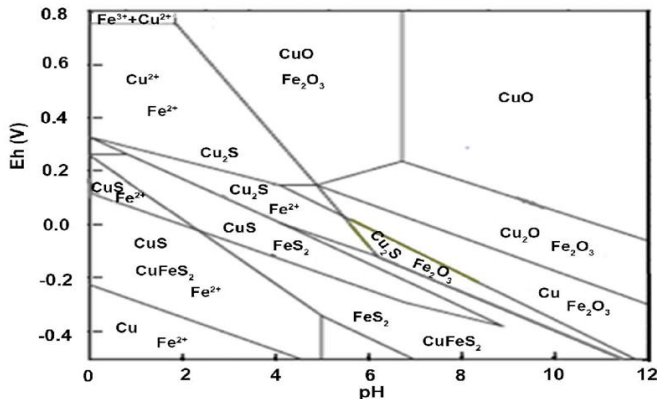


Fig. 5: Pourbaix diagram for Cu-Fe-S H₂O system

The metallurgical information drawn from the observations in figure above, is such that acidity is a key parameter for the dissolution of copper during leaching. Further looking at the Pourbaix diagram it is evident that Copper dissolve in a solution at a Ph ranging from approximately 1-2 at an Eh of about 0,3-0,7 V, comparing these conditions with the working conditions used in this experiment which were pH of 1.5 at a potential of approximately 0.31V, the low dissolution of copper might be justified by the reason that the phases that occur at the optimal conditions which are Cu^{2+} , Fe^{2+} and Cu_2S . It is shown in the diagram above that the dominant phase at the condition that was used in this experiment is Cu_2S which might have passivated on the surface of chalcopyrite thus leading to low dissolution rate of Cu. This therefore lowered the recovery of Cu in the leaching solution and further decrease the release of Cu ions.

Based on the experiment conducted by Nazari et al [17], galena does not increase the dissolution rate of copper instead it retards it. According to the results obtained, they agree with the findings of Nazari et al [17]. Comparing the rest potential of both PbS and CuFeS_2 , 0.4V and 0.56V, respectively it can be deduced that if there was galvanic interaction galena is the one that underwent oxidation and chalcopyrite was the cathode. One of the reasons for the dissolution of Cu to be

slow when CuFeS_2 is coupled with PbS , it might be because of the possible dissolution of PbS which might contribute to the increase of sulphur content into the solution therefore the layer of sulphur reduced the surface area of CuFeS_2 exposed to the leaching solution and make it more difficult for H^+ ions to access and react with the chalcopyrite surface, slowing down copper dissolution.

D. Effects of Temperature

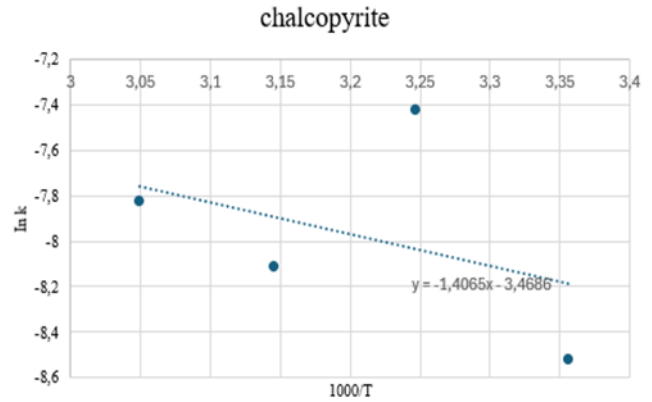


Fig. 6: Arrhenius plot obtained for chalcopyrite dissolution

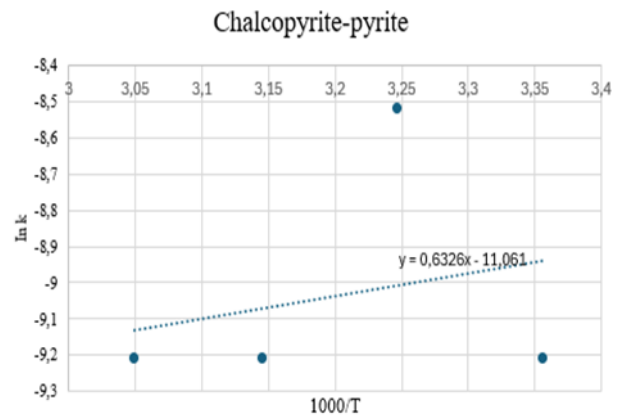


Fig. 7: Arrhenius plot obtained for chalcopyrite-pyrite

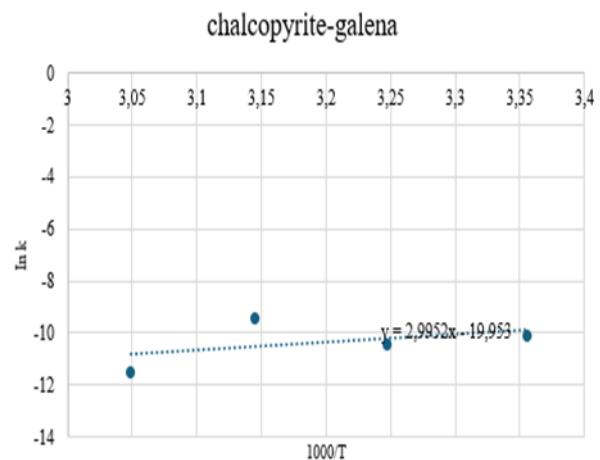


Fig. 8: Arrhenius plot obtained for chalcopyrite-galena

It was discovered that temperature has a positive impact on

recovery and rate. Therefore, when surface chemical reaction regulates, its influence was assessed using the shrinking core model, as indicated in Equation (5).

$$1 - (1 - x)^{1/3} = kt$$

where a is the copper ion concentration, k corresponds to the reaction rate constant, and t means the leaching time. The Arrhenius equation was used to calculate the activation energy of chalcopyrite leaching:

$$\ln \left(\frac{1 - 3(1 - x)^{2/3} + 2(1 - x)}{k} \right) = \ln A - \frac{E_a}{RT}$$

$$k = Ae^{-E_a/RT}$$

where A is the pre-exponential factor, E_a is the activation energy (J·mol⁻¹), R is the universal gas constant (J·mol⁻¹·K⁻¹) and T is the absolute temperature in Kelvin (K).

The CuFeS₂ concentrate sample dissolution had the highest E_a (around 19.69364 kJ/mol). The activation energy decreased with the addition of PbS and FeS₂, being around -5,25944 kJ/mol and -24,9021 kJ/mol, respectively. Small activation energy values are related to the case where diffusion is rate-limiting step. However, different values, 12 - 26 KJ/mol, are reported for activation energy required for diffusion step [18]. Moreover, different values (40 -80 KJ/mol) are reported for activation energy where chemical reaction is the controlling factor. According to Karimov et al [19] to the results, the rate limiting step for the dissolution of CuFeS₂ was surface chemical reaction, while with addition of PbS or FeS₂ was found to be diffusion through product layer. However, the equation that gave the best fit for all 3 process was surface chemical reaction. Chalcopyrite's poor reactivity in its natural form is reflected in its high activation energy barrier for dissolution, which is determined by thermodynamic analysis. Even while galvanic interactions in the presence of pyrite somewhat lower this high energy barrier, it was still unable to promote extensive Cu dissolution.

IV. CONCLUSION

The aim of this experiment was to compare the impacts of galvanic couples which are CuFeS₂/FeS₂ and CuFeS₂/PbS on the kinetic dissolution of copper from chalcopyrite (CuFeS₂). According to the results, temperature has a major effect on the effectiveness of chalcopyrite leaching, with Cu dissolution rates rising with higher temperatures. After 4 hours at 55°C, chalcopyrite leached alone produced a high Cu recovery of almost 67%, demonstrating that high temperatures accelerate the dissolving process. Nevertheless, pyrite and galena unexpectedly decreased Cu recovery. The results showed that pyrite did not efficiently catalyse Cu dissolution, with the highest Cu recovery limited at 38% at 55°C, in contrast to literature that asserts pyrite should improve chalcopyrite leaching through galvanic interactions instead. Cu recovery was further reduced with the addition of galena, reaching a maximum recover of 13% at 55°C. Chalcopyrite's stability and resistance to dissolve are further highlighted by the activation energy analysis; a high E_a value (19.69 kJ/mol) suggests that a surface chemical reaction is the rate-limiting phase. The

introduction of galena and pyrite decreased this barrier, indicating a change to diffusion through the product layer. Nevertheless, the mixed mineral systems' Cu recovery was not considerably enhanced by the activation energy reduction.

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