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## SELECTIVE OXIDATION OF IRON IN CHALCOPYRITE FOR ENHANCED COPPER RECOVERY

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**SELECTIVE OXIDATION OF IRON IN CHALCOPYRITE FOR ENHANCED COPPER RECOVERY**

KH.SH.SULTONOV, SH.T.KHOJIEV, G.B.BEKNAZAROVA, M.S.SAIDOVA (Tashkent State Technical University, Republic of Uzbekistan)\*

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**Abstract:** This study explores the thermodynamic feasibility of selectively oxidizing iron contained in chalcopyrite mineral while preventing the oxidation of copper sulphide, with the ultimate goal of enhancing the copper concentration in the sulphide concentrate. A redox reaction in the solid phases between chalcopyrite and copper (I) oxide was constructed and subjected to thermodynamic analysis. The research identifies a crucial temperature range of 498-598 K (225-325 °C) as the most favourable for the redox reaction. The equilibrium constants at the selected optimal temperatures, 1.152 for chalcopyrite and 1.137 for copper (I) oxide, indicate that the redox reaction adheres to the expected kinetic behaviour. The significance of these findings lies in the potential to selectively oxidize iron in chalcopyrite, allowing for its subsequent removal through magnetic separation. This strategic approach promises an increase in the copper percentage within the sulphide concentrate, thereby enhancing the efficiency and economic viability of copper extraction processes. The study not only provides valuable insights into the thermodynamics of the chalcopyrite-copper (I) oxide redox reaction but also establishes a practical temperature range for its optimal execution. The successful implementation of this approach holds considerable promise for the mining and metallurgical industries, offering a pathway to improve copper recovery processes and addressing the economic challenges associated with traditional extraction methods.

**Keywords:** Chalcopyrite, Copper (I) oxide, Oxidation, Thermodynamics, Mechanism, Optimal temperature, economic.

**Annotatsiya:** Ushbu tadqiqotda mis sulfidining oksidlanishini oldini olish bilan xalkopirit mineralidagi temirni selektiv oksidlashning termodinamik maqsadga muvofiqligi o'rganilgan, buning asosiy maqsadi sulfidli mis boyitmasida mis konsentratsiyasini oshirishdir. Xalkopirit va mis (I)-oksidi orasidagi qattiq fazadagi oksidlanish-qaytarilish reaksiyasi tuzilgan va termodinamik tahlildan o'tkazilgan. Tadqiqot 498-598 K (225-325 °C) kritik harorat oralig'ini oksidlanish-qaytarilish reaksiyasi uchun eng maqbul sharoit deb topilgan. Tanlangan optimal haroratlarda muvozanat konstantalari xalkopirit uchun 1,152 va mis (I)-oksidi uchun 1,137 bo'lgan oksidlanish-qaytarilish reaksiyasi kutilgan kinetik natijalardan dalolat beradi. Ushbu natijalarning ahamiyati temirning xalkopiritdagi selektiv oksidlanish imkoniyatidadir, bu esa uni keyinchalik magnitli usulda ajratish orqali olib tashlash imkonini beradi. Ushbu strategik yondashuv sulfidli boyitma tarkibidagi misning massa ulushini oshirishga va shu orqali misni qayta ishlash jarayonlarining unumdorligi hamda iqtisodiy samaradorligini oshirishga imkon beradi. Tadqiqot nafaqat xalkopirit-mis (I)-oksid orasidagi oksidlanish-qaytarilish reaksiyasining termodinamikasi haqida qimmatli ma'lumotlarni beradi, balki uning optimal ishlashi uchun amaliy harorat oralig'ini ham belgilab beradi. Ushbu yondashuvning muvaffaqiyatli amalga oshirilishi tog'-kon va metallurgiya sanoati uchun muhim jihatga ega bo'lib, mis qazib olish jarayonlarini takomillashtirish va an'anaviy metallurgiya usullari bilan bog'liq iqtisodiy muammolarni hal qilish yo'lini taklif qiladi.

**Tayanch so'zlar:** xalkopirit, mis (I)-oksidi, oksidlanish, termodinamika, mexanizm, optimal harorat, iqtisod.

**Аннотация:** В этом исследовании изучается термодинамическая осуществимость селективного окисления железа, содержащегося в минерале халькопирит, при одновременном предотвращении окисления сульфида меди, с конечной целью повышения концентрации меди в сульфидном концентрате. Построена и подвергнута термодинамическому анализу окислительно-восстановительная реакция в твердых фазах между халькопиритом и оксидом меди (I). Исследование определило критический диапазон температур 498–598 K (225–325 °C) как наиболее благоприятный для окислительно-восстановительной реакции. Константы

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равновесия при выбранных оптимальных температурах 1,152 для халькопирита и 1,137 для оксида меди (I) указывают на то, что окислительно-восстановительная реакция соответствует ожидаемому кинетическому поведению. Значимость этих результатов заключается в возможности избирательного окисления железа в халькопирите, что позволяет его последующее удаление посредством магнитной сепарации. Этот стратегический подход обещает увеличение процентного содержания меди в сульфидном концентрате, тем самым повышая эффективность и экономическую целесообразность процессов извлечения меди. Исследование не только дает ценную информацию о термодинамике окислительно-восстановительной реакции халькопирит-оксид меди (I), но также устанавливает практический температурный диапазон для ее оптимального проведения. Успешная реализация этого подхода открывает значительные перспективы для горнодобывающей и металлургической промышленности, предлагая путь к улучшению процессов извлечения меди и решению экономических проблем, связанных с традиционными методами добычи.

**Ключевые слова:** халькопирит, оксид меди (I), окисление, термодинамика, механизм, оптимальная температура, экономика.

### Introduction

Copper, one of the most essential metals in modern society, finds its origins deeply embedded in the Earth's crust, existing in various compounds and native forms. With an average content in the Earth's crust ranging from 0.0047% to 0.0055%, copper remains a vital resource for numerous industries worldwide. Key industrial copper ores, including chalcopyrite ( $\text{CuFeS}_2$ ), chalcocite ( $\text{Cu}_2\text{S}$ ), and bornite ( $\text{Cu}_5\text{FeS}_4$ ), serve as primary sources for copper extraction. These ores are found in diverse geological formations across the globe, with notable deposits located in regions such as Udokan, Zhezkazgan, Mansfeld in Germany, and prominent sites in Chile and the USA [1, 2,3].

In the mineral-rich landscape of Uzbekistan, approximately 900 occurrences of copper ore have been identified, reflecting the country's significant potential in copper extraction. However, despite the abundance of copper ore occurrences, only a few hydrothermal porphyry deposits are actively exploited, primarily concentrated in the Almalyk region. Notable sites in this region include Kalmakyr, Dalnee, and Sary-cheku, with smaller deposits like Balykty and Karabulak contributing to the country's copper production. Economic viability in Uzbekistan's copper mining industry necessitates copper concentrations above 0.35%, driving exploration and extraction efforts towards high-grade deposits [4, 5, 6].

Pyrometallurgical methods remain the primary means of copper extraction in Uzbekistan, with chalcopyrite serving as a principal source ore due to its prevalence and copper content ranging from 0.5% to 2.0%. The extraction process typically involves flotation enrichment to obtain copper concentrate, which is then subjected to smelting in specialized furnaces. During smelting, excess sulphur is removed as gases (5-8%  $\text{SO}_2$ ), often utilized in the production of sulfuric acid ( $\text{H}_2\text{SO}_4$ ). Furthermore, smelting facilitates the conversion of impurities such as iron, zinc, arsenic, and lead into slag, which is separated from the molten copper. Various melting furnaces, including oxygen-flare, reverberatory, and electric

furnaces, among others, are employed to achieve efficient copper extraction from the concentrate [7,8, 9].

Despite its widespread occurrence and importance as a copper source, chalcopyrite presents challenges in traditional extraction methods due to its significant iron content. Smelting processes generate substantial amounts of slag, contributing to environmental concerns and reducing operational efficiency. To address these challenges, ongoing research proposes a novel approach centered on selectively oxidizing iron in chalcopyrite using copper (I) oxide [10,11].

The significance of this study lies in its potential to enhance the efficiency of copper recovery processes while concurrently minimizing slag formation during smelting operations. By targeting the selective oxidation and subsequent removal of iron from chalcopyrite, researchers aim to increase the copper content in the concentrate, thereby improving the overall economics of copper extraction. Moreover, this innovative approach aligns with sustainability goals within the metallurgical industry, as it offers the prospect of reducing waste generation and energy consumption associated with traditional smelting methods [12,13].

The research endeavours to explore the thermodynamic feasibility of selective oxidation processes and their practical applications in optimizing copper production. Through a combination of theoretical modelling, laboratory experimentation, and process optimization, the study aims to provide valuable insights into the development of cleaner, more efficient, and sustainable practices for copper extraction. Ultimately, the findings of this research hold the potential to revolutionize the copper mining industry by offering a pathway towards greener and more economically viable extraction processes.

### Research Methods and the Received Results

This research focuses on studying the interaction between the mineral chalcopyrite and

copper (I) oxide to understand the potential for copper enrichment. The main goal is to analyse the thermodynamic aspects of this interaction. The research methodology consisted of creating a model of the reaction of copper (I) oxide and chalcopyrite. Using the Thermo-Calc program, we determined the thermodynamic quantities associated with their chemical reaction. In addition, Microsoft Excel was used to generate an Ellingham plot, which provided valuable information about the thermodynamic behaviour of the process. This state-of-the-art analysis uses advanced tools to examine the potential and consequences of interactions between chalcopyrite and copper (I) oxide for copper enrichment.

Process of reaction of chalcopyrite with copper (I) oxide. Based on practical experiments and considering the chemical and mineralogical composition of the products obtained, a reliable general chemical equation for the reaction of chalcopyrite with copper (I) oxide (Cu<sub>2</sub>O) is as follows:



This chemical equation represents the equilibrium reaction, where chalcopyrite (CuFeS<sub>2</sub>) reacts with copper (I) oxide (Cu<sub>2</sub>O). The resulting products include copper sulphide (CuS), iron oxide (FeO), sulphur dioxide (SO<sub>2</sub>), and copper (Cu). This reaction is essential for understanding the composition and behaviour of the system in practical applications, shedding light on the equilibrium state reached during the interaction between chalcopyrite and copper (I) oxide [14].

Utilizing reference books, we calculated thermodynamic values for all substances involved in the reactions under standard conditions, establishing an initial database for the reaction. The compiled data are presented in Table 1. Building upon this foundation, we have formulated new applications and incorporated additional information to further enhance the understanding of the composition and thermodynamics of the reaction. The expanded dataset serves as a valuable resource for exploring the intricacies of the chalcopyrite-copper (I) oxide reaction and its thermodynamic implications.

Table 1

Corresponding thermodynamic quantities of substances (at 298 K)

Thermodynamic parameters	CuFeS <sub>2</sub>	Cu <sub>2</sub> S	FeO	SO <sub>2</sub>	CuS	Cu <sub>2</sub> O
ΔH° (kJ/mol)	-188.3	-79.5	-272	-296.9	-53.1	-173.18
ΔG° (kJ/mol)	-194.1	-86.2	-251.46	-300.21	-53.6	-150.54
ΔS° (J/(mol*K))	143.8	120.9	60.75	248	66.5	92.93

Thermodynamic analysis of the chemical reaction of chalcopyrite and copper (I) oxide. Constant pressure heat capacity (C<sub>p</sub>) is a vital thermodynamic property that elucidates how the enthalpy of a substance changes with temperature under constant pressure conditions. While C<sub>p</sub> is universally applicable, the specific method of calculation may vary based on the substance's nature (ideal or real gas, solid, liquid) and the available data. The experimental determination of C<sub>p</sub> for a single substance is crucial and often dependent on a range of temperatures. When dealing with a system comprising diverse substances undergoing reactions, the computation of C<sub>p</sub> for the entire system becomes more intricate. The total heat capacity of the system is the aggregate of individual heat capacities of reactants and products, each multiplied by their respective stoichiometric coefficients. This system-level analysis provides a comprehensive understanding of the thermodynamics at play during reactions, shedding light on how the enthalpy changes with temperature and pressure throughout the entire process.

Mathematically, the total heat capacity (C) of a system can be expressed as the sum of the products of the stoichiometric coefficients (v<sub>i</sub>) and the heat capacities at constant pressure (C<sub>p</sub>) for each particle (i) participating in the reaction:

$$C = \sum v_i C_{p,i} \quad (2)$$

Here C<sub>p,i</sub> represents the heat capacity at constant pressure for particle i, and v<sub>i</sub> is the stoichiometric coefficient of particle i in the reaction.

This mathematical representation encapsulates the combined impact of individual particles on the overall heat capacity of the system. The change in enthalpy (ΔH) for the reaction is determined by the following formula:

$$\Delta H_{\text{reaction}} = \int_{T_1}^{T_2} C_p dT \quad (3)$$

Assuming that the heat capacity at constant pressure (C<sub>p</sub>) remains constant over the temperature range, the integral simplifies to ΔC<sub>p</sub>ΔT.

This mathematical framework allows for a quantitative understanding of the heat capacity contributions from each particle involved in the reaction, providing a foundation for further thermodynamic analyses and system optimizations.

The change in enthalpy of a reaction is determined by the following mathematical formula (ΔH):

$$\Delta H = \Delta H^0 + \int_{T_1}^{T_2} C_p dT \quad (4)$$

Here: C<sub>p</sub> is the heat capacity at constant pressure. If it is constant over the entire temperature range, the integral simplifies to C<sub>p</sub>:



$$\Delta H = \Delta H^0 + C_p \Delta T \quad (5)$$

And the change in enthalpy of the reaction ( $\Delta S$ ):

$$\Delta S = \Delta S^0 + \int_{T_1}^{T_2} \frac{C_p}{T} dT \quad (6)$$

Again, assuming  $C_p$  is constant, the integral simplifies to  $C_p/T\Delta T$ .

$$\Delta S = \Delta S^0 + \frac{C_p}{T} \Delta T \quad (7)$$

Gibbs free energy change ( $\Delta G$ ):

$$\Delta G = \Delta H - T\Delta S \quad (8)$$

Substituting the above expressions ( $\Delta H$ ) and ( $\Delta S$ ):

$$\Delta G = (\Delta H^0 + C_p \Delta T) - T(\Delta S^0 + \frac{C_p}{T} \Delta T) \quad (9)$$

Depending on the value of the Gibbs energy, systems make a conclusion about whether reactions will proceed spontaneously or not, and about the direction of the reaction at a given temperature:

a) if the magnitude of the change in the Gibbs energy during a chemical reaction is  $\Delta G_T > 0$ , positive, then the reaction cannot proceed spontaneously at a given temperature;

b) if  $\Delta G_T < 0$ , negative, then the reaction proceeds spontaneously at a given temperature. Then this reaction is cost-effective and technologically convenient at a given temperature;

d) if  $\Delta G_T = 0$  then the system is in a state of chemical equilibrium. It is necessary to change the system parameters ( $P$ ,  $T$ ,  $C$ ,  $V$ ) to direct the balance towards the required reaction [5].

The temperature dependence of the reaction equilibrium constant ( $K_e$ ) was determined using the following formula:

$$\Delta G = -RT \ln K_e \quad (10)$$

Here:  $R$  – universal gas constant,  $R = 8.31696 \cdot 10^{-3}$  kJ/(deg·mol);

$K_e$  is the equilibrium constant of the corresponding chemical reaction.

From the mathematical expression (8), the equilibrium constant is equal to:

$$K_e = e^{-\frac{\Delta G}{RT}} \quad (11)$$

Thermodynamic values of all substances participating in the reactions are calculated according to Hess's law for standard conditions, and their initial values are given in Table 2.

Table 2  
Values of the reaction of chalcopirite with copper (I) oxide under standard conditions (298 K)

No	$\Delta H^0$ (kJ/mol)	$\Delta S^0$ (kJ/(mol·K))	$\Delta G^0$ (kJ/mol)
1	-322.34	532.86	-377.51

The values presented in Table 3 mean that the reaction of chalcopirite with copper (I) oxide is exothermic, that is, the oxidation of 1 mole of chalcopirite by reaction releases -322.34 kJ of heat under standard conditions. The fact that the reaction is exothermic increases the importance of this process and significantly reduces fuel consumption when used in industry [15,16].

The Gibbs energy value calculated for standard conditions was also negative. So, in accordance with the laws of thermodynamics, we can conclude that this reaction proceeds spontaneously under standard conditions. But given that this reaction occurs in the solid phase, to start the reaction it is enough to provide the initial activation energy. Additionally, the favourable thermodynamic conditions suggest that this reaction can occur readily without the need for external energy input, as the system inherently possesses the necessary energy to overcome the activation barrier [17].

Such reactions play a pivotal role in the metallurgical sector, where the energy liberated in the initial collision acts as the catalyst for subsequent collisions. This cascade effect leads to the formation of chain reactions within the process, driving efficiency and productivity in metallurgical operations [18,19].

The reaction entropy value, measured at 532.86 kJ/(mol K) under standard conditions, highlights an increase in the degree of particle disorder. This phenomenon stems from the formation of sulphur dioxide as a by-product of the reaction, contributing to the overall entropy change. Moreover, the release of gaseous sulphur dioxide molecules further enhances the system's disorderliness, reflecting the spontaneity of the reaction [20].

Table 3

Changes in the Gibbs energy and equilibrium constant with increasing temperature in the reaction of chalcopirite with copper (I) oxide

No	T, K	T, °C	$\Delta G^T$ , kJ/mol	$K_e$
1	2	3	4	5
1	298	25	-377.51	1.16467758
2	348	75	-507.775	1.19194517
3	398	125	-534.418	1.17537068
4	448	175	-561.061	1.16265508
5	498	225	-587.704	1.15259178

1	2	3	4	5
6	548	275	-614.347	1.14442959
7	598	325	-640.990	1.13767645
8	648	375	-667.633	1.13199656
9	698	425	-694.276	1.12715298
10	748	475	-720.919	1.12297369
11	798	525	-747.562	1.11933082
12	848	575	-774.205	1.11612735
13	898	625	-800.848	1.11328831
14	948	675	-827.491	1.11075487
15	998	725	-854.134	1.1084802
16	1048	775	-880.777	1.1064266
17	1098	825	-907.420	1.10456332
18	1148	875	-934.063	1.1028651
19	1198	925	-960.706	1.10131092
20	1248	975	-987.349	1.09988321
21	1298	1025	-1013.99	1.09856714
22	1348	1075	-1040.63	1.0973501
23	1398	1125	-1067.27	1.09622132
24	1448	1175	-1093.92	1.09517154
25	1498	1225	-1120.56	1.09419275

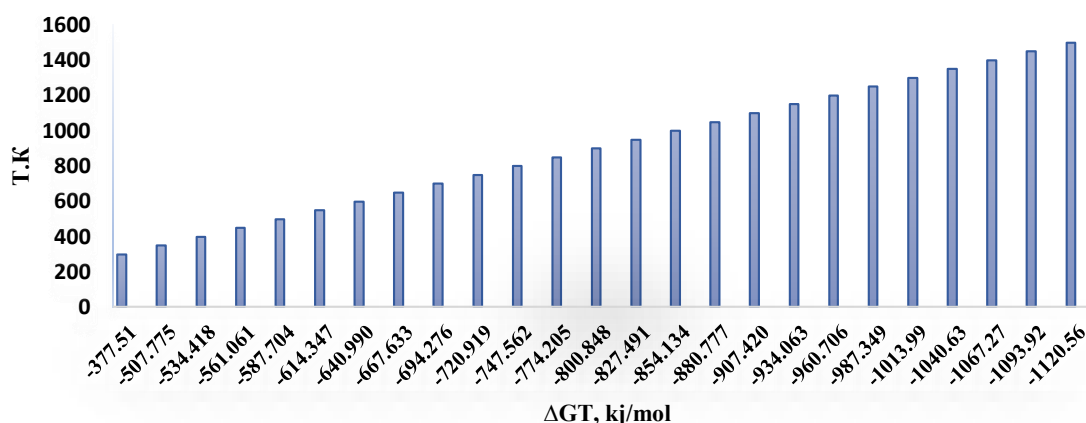


Fig. 1. Change in the Gibbs energy of the system with increasing temperature in the reaction of chalcopyrite and copper (I) oxide

From the values of the table 3 it can be seen that the Gibbs energy of the reaction system decreases with increasing temperature. This indicates that increasing the temperature in the system increases the likelihood of a reaction (Figure 1).

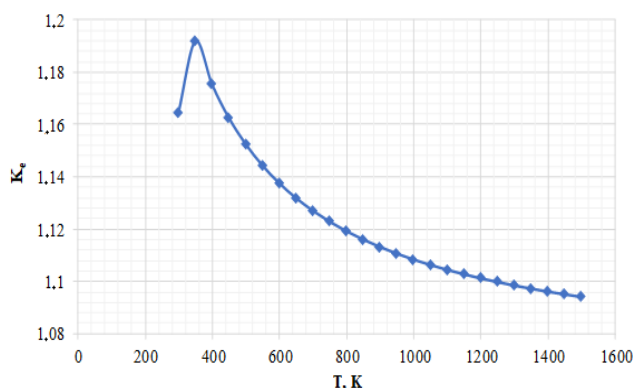


Fig.2. Change in the equilibrium constant during the oxidation of chalcopyrite with copper (I) oxide depending on temperature

However, it was found that with increasing temperature the equilibrium constant increases from 298 K (25 °C) to 348 K (75 °C), and above 348 K (75 °C) it decreases (Fig. 2).

In practice, taking into account the process of oxidation of chalcopyrite with copper (I) oxide, the radiation of heat into the environment when it is carried out in a furnace, the temperature range is 498-598 K (225-325 °C). Can be chosen as the optimal condition. Because at a temperature below 498 K, the heat supplied to the furnace will be difficult to be completely absorbed by the solid raw material mixture. This situation is explained by the heat consumption from the furnace walls and flue gases. An increase in temperature above 598 K leads to a decrease in the equilibrium constant of the reaction.

### Conclusion

From a thermodynamic point of view, the possibility of selective oxidation of iron contained in the chalcopyrite mineral by copper (I) oxide is

considered. In this case, it was necessary to prevent the oxidation of copper sulphide. The importance of taking this condition into account is that if the iron in chalcopyrite is selectively oxidized and then removed by magnetic separation, an increase in the percentage of copper in the copper sulphide concentrate is achieved.

To fulfil the above requirements, the redox reaction between chalcopyrite and copper (I) oxide in the solid phase was constructed and the thermodynamics were analysed. As a result of thermodynamic analysis, the temperature range of 498-598 K (225-325 °C) was selected as the most favourable (optimal) temperature for carrying out the redox reaction between chalcopyrite and copper (I) oxide. It has been established that the equilibrium constant of the redox reaction between chalcopyrite and copper (I) oxide in the range of temperatures chosen as optimal is 1.152 and 1.137, respectively. These indicators indicate that the reaction is proceeding as expected from a kinetic point of view.

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