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Metallurgy

Computer simulation of the interaction of copper monosulfide with sodium chloride in the presence of boron trioxide

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ABSTRACT

Expansion of the raw material base of ferrous metallurgy depends to some extent on the creation of an effective technology for the complex processing of copper-magnetite ores with copper extraction. The article presents the results of studies of copper chlorination from CuS present in sulfide-magnetite ores using sodium chloride and boron trioxide. The studies were carried out in the temperature range of 500-1500 °C and a pressure of 0.1-0.001 bar by thermodynamic modeling using the HSC-10 software package based on the principle of minimum Gibbs energy. It was found that the interaction in the $CuS-NaCl-B_2O_3-O_2$ system occurs with the formation of copper chlorides (Cu_4Cl_4, Cu_3Cl_3, Cu_2Cl_2, CuCl), sodium borates ($Na_2B_4O_6$, $Na_2B_6O_{10}$), Na_2SO_4, $SO_{2(g)}$. The temperature of the maximum (89-90%) degree of copper extraction into gaseous chlorides decreases from 1050 to 850°C with a decrease in pressure from 0.1 to 0.001 bar. It was found that the chloride sublimation of copper is accompanied by the formation of elemental copper and gaseous NaCl during the interaction of Cu_3Cl_3 with $Na_2B_4O_6$.

Keywords: chalcopyrite-magnetite ore, thermodynamic modeling, temperature, pressure, chloride sublimation, copper.

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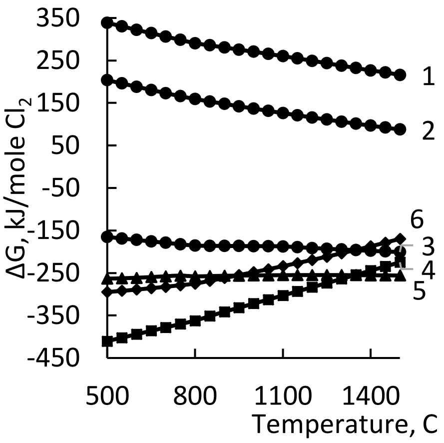
Introduction

As a rule, magnetite ores containing usually 55-60% iron due to the high magnetic susceptibility of the magnetite mineral [1] are subjected to magnetic concentration before metallurgical processing. This method of ore preparation is widely used and is constantly improved [[2], [3], [4], [5], [6], [7], [8]]. Problems with processing magnetite ores arise in the case of the presence of non-ferrous metals in the ore, including copper. Special technologies are developed for copper extraction from magnetite ores. Thus, in [9] a flotation-magnetic technology for complex processing of copper-containing sulfide-magnetite ore is described. The technology allows extracting 87.1-91.7% copper into concentrate, with a content of 19.1-21.6% of this metal. 84.3-87.9% of iron is extracted into iron concentrate (66.5% Fe). For processing magnetite ore containing 0.093% copper, a multistage magnetic flotation technology was developed [[10], [13]]. At the first stage, a concentrate containing 65-66% iron and tailings was obtained using the wet magnetic separation method. Copper is extracted from the tailings by flotation into a concentrate containing 15.2% Cu, 26.5% Fe, 17.5% S, 8.5% Zn, 16.2% Si. Multistage roller grinding with integrated magnetic separation stages between the stages is also used [14], which also allows iron to be extracted into a magnetic concentrate.

Iron-containing raw materials with impurities of non-ferrous metals include pyrite cinders [15], blast furnace and steelmaking dust [16]. For selective separation of iron from non-ferrous metals from this raw material with a concentration of 30-66% iron, chloride sublimation processes using gaseous and solid chlorinating agents are used in metallurgical practice [15]. The copper extraction degree into sublimes in this case is 92%, lead – 95%, zinc and silver – 97%. The cinder after chloride sublimation roasting is of interest to ferrous metallurgy plants. The most common chlorinating agents in terms of decreasing reactivity form a series: Cl_2, HCl, NH_4Cl, $CaCl_2$, NaCl, $NaCl \cdot MgCl_2$ [17]. The disadvantage of using Cl_2 and HCl is toxicity. Calcium chloride is most widely used in chloride sublimation. However, the price of technical calcium chloride is 190 KZT/kg [18]. Less expensive is technical sodium chloride. Its price is 43 KZT/kg [19]. However, it is inferior in reactivity to calcium chloride [17]. The reactivity of the widely used and relatively cheap NaCl can be increased if substances are formed in the chlorination products, the ΔG° of which is more negative than the total ΔG° of the original substances. For this purpose, a calculation of the ΔG° of chloride sublimation of CuS (present in magnetite ores) with sodium chloride was carried out to form Na_2SO_4, SO_2, Na_2SiO_3, sodium borates ($Na_2B_4O_7$ and $Na_2B_6O_{10}$). The calculation was carried out using the HSC-10 complex [20].



The numbers on the lines correspond to the reaction numbers

Figure 1 – Influence of temperature on the ΔG° of the CuS chlorination reactions

The reactions under consideration:

$2NaCl + CuS = CuCl_{2(g)} + Na_2S$; (1)

$CaCl_2 + CuS = CaCl_{2(g)} + CaS$; (2)

$2NaCl+CuS+SiO_2+1.5O_2 = CuCl_2+Na_2SiO_3+SO_2$; (3)

$2NaCl + CuS +2O_2 = CuCl_{2(g)} + Na_2SO_4$; (4)

$2NaCl+CuS+2B_2O_3+1.5O_2=CuCl_{2(r)}+Na_2B_4O_7+SO_2$; (5)

$2NaCl+CuS+3B_2O_3+1.5O_2=CuCl_{2(g)}+Na_2B_6O_{10}+SO_2$. (6)

The influence of temperature on the ΔG° of the reactions is shown in Figure 1.

From the given ΔG° values it is seen that at a temperature >1300°C the reactivity of NaCl is the highest if the CuS chloride sublimation is carried out in the presence of B_2O_3 and oxygen with the formation of $Na_2B_4O_7$. The process can be carried out in the presence of oxygen (reaction 4). However, in this case sulfur remains in the cinder in the form of Na_2SO_4, which is undesirable for further use of the cinder in ferrous metallurgy. Based on this, further studies were carried out using the method of computer thermodynamic modeling of the interaction in the CuS- CuS-NaCl-B_2O_3-O_2 system.

Experimental part

Thermodynamic modeling of the process was carried out using the HSC-10 Chemistry software package developed by the Finnish metallurgical company Outokumpu [16]. The calculation of equilibrium using the HSC-10 software package is based on the principle of minimum Gibbs energy taking into account the activities of substances (Equilibrium Compositions subprogram). The minimum ΔG° is found using the Lagrange functions and the Newton method of successive approximations. The HSC-10 program database contains information on the enthalpy of formation, entropy, heat capacity, heats of phase and modification transitions of 18 000 substances. According to the program developers, it is very difficult to obtain the absolute calculation error value. Nevertheless, based on the fact that the program database is constantly reviewed and refined, the calculation functions of the HSC software package are quite reliable, and the results are adequate [20].

The influence of temperature from 500 to 1500°C and pressure from 0.001 to 0.1 bar on the equilibrium degree of chlorination and chloride sublimation of copper and the behavior of boron, sodium and sulfur were determined.

Results and Discussion

Figure 2 shows the equilibrium distribution degree (α, %) of copper, boron, sodium and sulfur at a pressure of 0.1 bar.

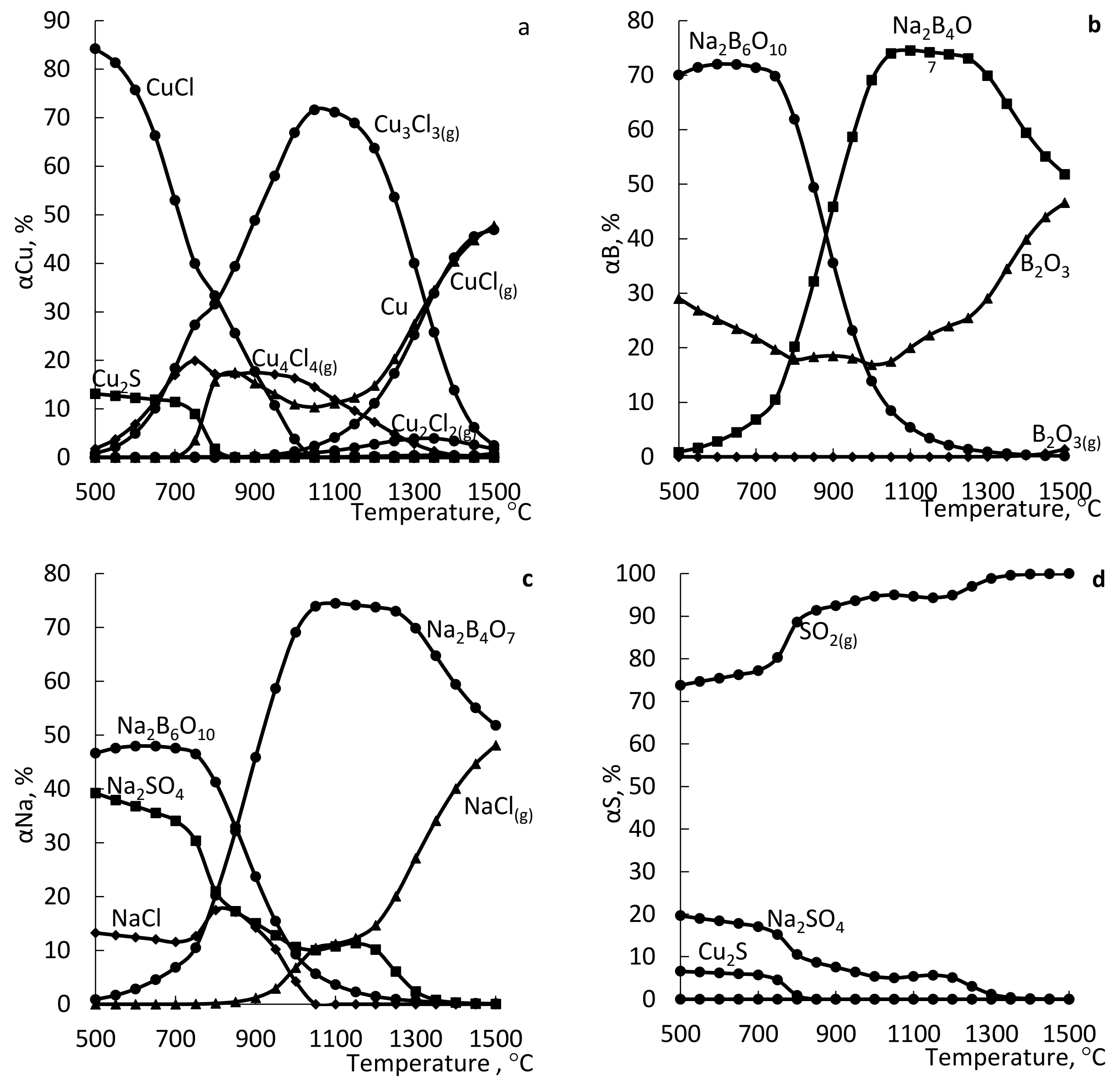


Figure 2 – Influence of temperature on the equilibrium degree of distribution of copper (a), boron (b), sodium (c) and sulfur (d)

It is seen that the main products in the system under consideration are $Cu_4Cl_{4(g)}$, $Cu_3Cl_{3(g)}$, $Cu_2Cl_{2(g)}$, $CuCl_{(g)}$, $CuCl_2$, Cu_2S, Cu, $Na_2B_6O_{10}$, $Na_2B_4O_7$, Na_2SO_4, $SO_{2(g)}$, $NaCl_{(g)}$.

At 500°C, most (84.2%) copper is chlorinated to CuCl. At a temperature above 850°C, gaseous $Cu_4Cl_{4(g)}$, $Cu_3Cl_{3(g)}$ and $Cu_2Cl_{2(g)}$ begin to form. An undesirable process is the formation of elemental copper at a temperature above 700°C. The formation of gaseous monochloride – CuCl occurs at a temperature above 700°C. Moreover, this process develops at a temperature above 1050°C.

A small part of copper (13.1%) at 500°C passes from CuS to Cu_2S, which disappears at 1050°C. Initially, at 500°C, the main part of boron (70%) is in $Na_2B_6O_{10}$. Then the transition of boron to $Na_2B_6O_{10}$ decreases, and in $Na_2B_4O_7$ it increases (up to 73.5% at 1050-1150°C). It should be noted that in the system at a temperature of >1100°C the boron transition degree to B_2O_3 increases significantly. Most sulfur in the temperature range of 500-1500°C passes from CuS to SO_2.

At 500°C, the main part of sodium (46.6%) passes from NaCl to $Na_2B_6O_{10}$ and $NaSO_4$ (39.2%). Sodium chloride does not react completely. 13.2% of it did not react. Gaseous NaCl appears at a temperature above 800°C. At 1200°C, this process develops significantly.

The appearance of Cu_2S in the system is explained by the reaction:

$$2CuS + O_2 = Cu_2S + SO_2, \quad (7)$$

the ΔG° of which at 500°C is -291.7 kJ, and the formation of Na_2SO_4 is explained by the reaction:

$2CuS + 2NaCl + O_2 = Na_2SO_4 + 2CuCl + 2SO_2$, (8)

the ΔG° of which at 500°C is -787.2 kJ.

As can be seen from Figure 2, in the system under consideration, copper chlorides form a homologous series from $Cu_4Cl_{4(g)}$ to $CuCl_{(g)}$. Figure 3 shows the thermal behavior of $Cu_4Cl_{4(g)}$. As the temperature increases, $Cu_4Cl_{4(g)}$ transforms into $Cu_3Cl_{3(g)}$ ($3Cu_4Cl_{4(g)} \rightarrow 4Cu_3Cl_{3(g)}$), which then decomposes into $CuCl_{(g)}$ ($Cu_3Cl_{3(g)} \rightarrow 3CuCl_{(g)}$).

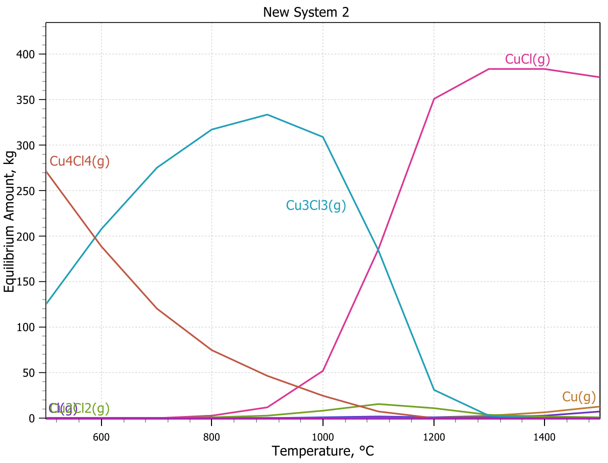


Figure 3 – Thermal behavior of gaseous $Cu_4Cl_{4(g)}$

From the comparison of the behavior of $Na_2B_4O_7$, $NaCl_{(g)}$, B_2O_3, Cu_3Cl_3, CuCl, Cu, and oxygen, it is seen that with an increase in temperature above 1050°C, the copper transition Cu_3Cl_3 decreases, while it increases in $CuCl_{(g)}$ and Cu. At that, the sodium transition degree to $Na_2B_4O_7$ decreases, while it increases in $NaCl_{(g)}$. Figure 4 shows the interaction between $Na_2B_4O_7$ and $Cu_3Cl_{3(g)}$ according to the reaction:

$Na_2B_4O_7+2Cu_3Cl_{3(g)}=4CuCl_{(g)}+2NaCl_{(g)}+2B_2O_3+2Cu+0{,}5O_{2(g)}$. (9)

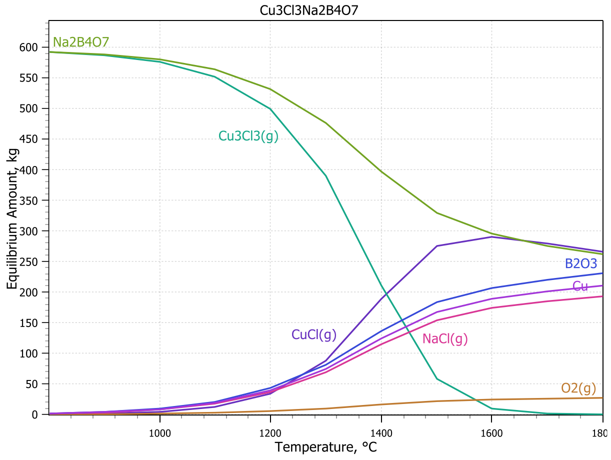


Figure 4 – Influence of temperature on the quantitative distribution of substances in the $Na_2B_4O_7 – 2Cu_3Cl_{3(g)}$ system at a pressure of 0.1 bar

It is seen that at 1000°C the interaction between $Na_2B_4O_7$ and $Cu_3Cl_{3(g)}$ occurs with the formation of copper, $CuCl_{(g)}$, B_2O_3 and oxygen. The possibility of copper formation from its chloride in the presence of $Na_2B_4O_7$ has not been described by anyone before. It was found by the article authors for the first time.

The obtained equilibrium values of the distribution of elements in the CuS-$NaCl$-B_2O_3-O_2 system allow to show the most probable step-by-step picture of the interaction using chemical equations.

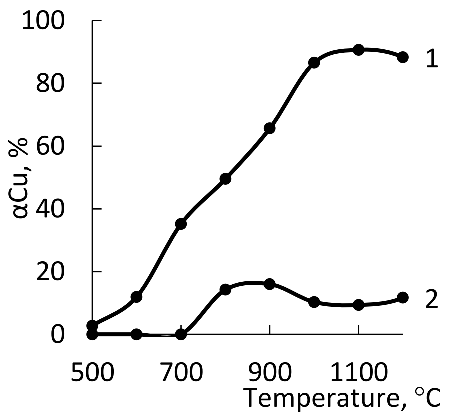
At 500°C: $2B_2O_3 + 2CuS + 2NaCl + 2{,}5O_{2(g)} = 0.58B_2O_3 + 0.01Na_2B_4O_7 + 0.47Na_2B_6O_{10} + 0.13Cu_2S + 0.01Cu_3Cl_{3(g)} + 0.01Cu_4Cl_{4(g)} + 1.68CuCl + 0.39Na_2SO_4 + 0.26NaCl + 1.48SO_{2(g)}$ (10)

At 750°C: 10th reaction products = $0.39B_2O_3 + 0.10Na_2B_4O_7 + 0.47Na_2B_6O_{10} + 0.07Cu + 0.09Cu_2S + 0.18Cu_3Cl_{3(g)} + 0.10Cu_4Cl_{4(g)} + 0.80CuCl + 0.30Na_2SO_4 + 0.25NaCl + 1.61SO_{2(g)}$ (11)

At 1050°C: 11th reaction products = $0.35B_2O_3 + 0.74Na_2B_4O_7 + 0.06Na_2B_6O_{10} + 0.21Cu + 0.01Cu_2Cl_{2(g)} + 0.48Cu_3Cl_{3(g)} + 0.07Cu_4Cl_{4(g)} + 0.05CuCl_2 + 0.10Na_2SO_4 + 0.21NaCl_{(g)} + 1.90SO_{2(g)}$ (12)

At 1300°C: 12th reaction products = $0.58B_2O_3 + 0.70Na_2B_4O_7 + 0.01Na_2B_6O_{10} + 0.55Cu + 0.04Cu_2Cl_{2(g)} + 0.27Cu_3Cl_{3(g)} + 0.01Cu_4Cl_{4(g)} + 0.51CuCl_2 + 0.01CuCl_{2(g)} + 0.02Na_2SO_4 + 0.54NaCl_{(g)} + 0.12O_{2(g)} + 1.98SO_{2(g)}$ (13)

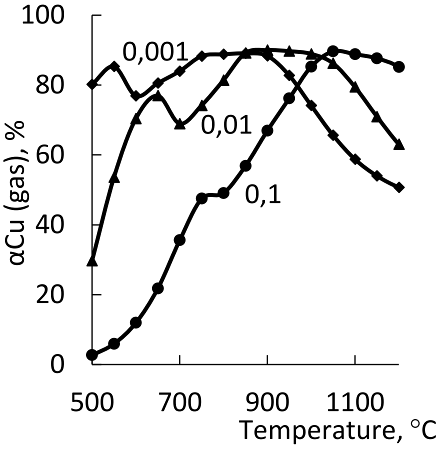
A rather complex relationship is observed between the total extraction of copper into the gas phase (in the form of ∑ chlorides) – $\alpha Cu_{(gas)}$ and temperature. From Figure 5 it is seen that the curve αCu(gas) = f(T) has a minimum, which is associated with the maximum formation of elemental copper.



The numbers on the line are pressure, bar

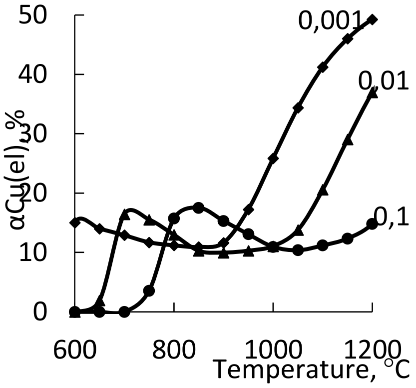
Figure 5 – Influence of temperature on the copper extraction degree into gaseous chlorides (1) and the degree of formation of elemental copper (2) at 0.1 bar

According to the Le Chatelier's principle, the equilibrium of reactions which products are gaseous substances can be shifted to the right by reducing the pressure in the system. Figure 6 shows the influence of pressure on the copper extraction degree into gaseous chlorides – $\alpha Cu_{(gas)}$.



The numbers on the line are pressure, bar

Figure 6 – Influence of temperature and pressure on the copper extraction into the gas phase



The numbers on the line are pressure, bar

Figure 7 – Influence of temperature and pressure on the elemental copper formation degree

From Figure 6 it is seen that the maximum degree of copper chloride sublimation (89.6-90%) decreases from 1050 to 850°C with a decrease in pressure from 0.1 to 0.01 bar in accordance with the equation:

$$T_{max} = 55.302 - 10.241 \bullet lgP - 37.677 \bullet P \qquad (14)$$

A decrease in pressure from 0.1 to 0.01 bar shifts the maximum (15-17%) formation of elemental copper to the low-temperature zone from 850 to 600°C (Fig. 7). It is seen that the observed maxima of copper chloride sublimation are associated with the minimum of elemental copper formation in these temperature zones.

At temperatures above 1000°C, a decrease in pressure increases the elemental copper formation degree. Thus, at 1200°C, a decrease in pressure from 0.1 to 0.001 bar leads to an increase in elemental copper formation from 14.5% to 49.5% in accordance with the equation:

$$\alpha_{Cu(el)} = 51.812 - 2.3874 \bullet lgP - 4.9051 \bullet lgP^2 \qquad (15)$$

Conclusions

Based on the obtained results on the interaction under equilibrium conditions in the $CuS\text{-}NaCl\text{-}B_2O_3\text{-}O_2$ system, the following conclusions can be drawn:

- the reactivity of NaCl with respect to CuS can be increased from a thermodynamic point of view if the reaction is carried out in the presence of B_2O_3 with the formation of sodium borates;
- at relatively low temperatures of 500-800°C, the reaction product is condensed CuCl and $Na_2B_6O_{10}$;
- chloride-containing products in the system are gaseous $Cu_5Cl_{5(g)}$, $Cu_4Cl_{4(g)}$, $Cu_3Cl_{3(g)}$, $Cu_2Cl_{2(g)}$, $CuCl_{(g)}$, $Cu_{(g)}$ and condensed Cu_2S, CuCl, Cu; boron, sodium, sulfur are present in the form of $Na_2B_4O_7$, $Na_2B_6O_{10}$, $NaCl_{(g)}$, Na_2SO_4, SO_2, B_2O_3;
- the temperature of the maximum degree of copper chloride sublimation at the level of 89.6-90.0% (in the form of ∑ copper chlorides) depends on the pressure, decreasing from 1050°C to 950°C with a decrease in pressure from 0.1 to 0.001 bar in accordance with equation: $T_{max}=55{,}302–10{,}241 \bullet lgP–37{,}677 \bullet P$; it is not possible to increase the degree of copper chloride sublimation from CuS due to the fact that sodium borate – $Na_2B_4O_7$ is not inert with respect to gaseous copper chlorides; when they interact (for example with Cu_3Cl_3), elemental copper and gaseous NaCl are formed.

Conflict of interest. On behalf of all the authors, the correspondent author declares that there is no conflict of interest.

CRediT author statement: **M. Tuleyev and V. Shevko:** Conceptualization; **V. Shevko:** Original Draft Preparation, Software; **M. Tuleyev, V. Shevko, D. Aitkulov:** Review & Editing; **M. Tuleyev and D. Aitkulov:** Visualization; **D. Aitkulov:** Project Administration.

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Бор (III) оксидінің қатысуымен мыс (II) сульфидінің натрий хлоридімен әрекеттесуін компьютерлік модельдеу

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ТҮЙІНДЕМЕ

Қара металлургияның шикізат базасын кеңейту белгілі бір дәрежеде мыс-магнетит кендерін мыс алу арқылы кешенді өңдеудің тиімді технологиясын құруға байланысты. Мақалада натрий хлориді мен бор (III) оксидін қолдана отырып, сульфид-магнетит кендерінде болатын CuS-тен мысты хлорлау зерттеулерінің нәтижелері келтірілген. Зерттеулер 500-1500°с температуралық интервалда және Гиббс минималды энергия принципіне негізделген HSC-10 бағдарламалық кешенін қолдана отырып, термодинамикалық модельдеу әдісімен 0,1-0,001 бар қысымда жүргізілді. CuS-$NaCl$-B_2O_3-O_2 жүйесіндегі өзара әрекеттесу мыс хлоридтерінің (Cu_4Cl_4, Cu_3Cl_3, Cu_2Cl_2, $CuCl$), натрий бораттарының ($Na_2B_4O_6$, $Na_2B_6O_{10}$), Na_2SO_4, $SO_{2(g)}$ түзілуімен жүретіні анықталды. Газ тәріздес хлоридтерге мысты экстракциялаудың максималды (89-90%) дәрежесінің температурасы қысымның 0,1-ден 0,001 барға дейін төмендегенде 1050-ден 850°С-қа дейін төмендейді. Мысты хлоридпен айдағанда (возгонка) Cu_3Cl_3 $Na_2B_4O_6$-мен әрекеттескенде мыс пен NaCl газының түзілуімен бірге жүретіні анықталды.

Түйін сөздер: халькопирит-магнетиті кені, термодинамикалық модельдеу, температура, қысым, хлорлы айдау, мыс.

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Компьютерное моделирование взаимодействия сульфида меди (II) с хлоридом натрия в присутвии оксида бора (III)

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АННОТАЦИЯ

Расширение сырьевой базы черной металлургии в некоторой степени зависит от создания эффективной технологии комплексной переработки медно-магнетитовых руд с извлечением меди. В статье приводятся результаты исследований хлорирования меди из CuS, присутствующего в сульфидно-магнетитовых рудах с использованием хлорида натрия и оксида бора (III). Исследования проводили в температурном интервале 500-1500°С и давлении 0,1-0,001 бар методом термодинамического моделирования с использованием программного комплекса HSC-10, основанного на принципе минимума энергии Гиббса. Найдено, что взаимодействие в системе CuS-$NaCl$-B_2O_3-O_2 происходит с образованием хлоридов меди (Cu_4Cl_4, Cu_3Cl_3, Cu_2Cl_2, $CuCl$), боратов натрия ($Na_2B_4O_6$, $Na_2B_6O_{10}$), Na_2SO_4, $SO_{2(g)}$. Температура максимальной (89-90%) степени извлечения меди в газообразные хлориды уменьшается от 1050 до 850°С при снижении давления от 0,1 до 0,001 бар. Найдено, что хлоридовозгонка меди сопровождается образованием элементной меди и газообразного NaCl при взаимодействии Cu_3Cl_3 с $Na_2B_4O_6$.

Ключевые слова: халькопирит-магнетитовая руда, термодинамическое моделирование, температура, давление, хлоридовозгонка, медь.

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