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STUDY OF THE THERMODYNAMIC PROPERTIES ABSORBATION SULFUR STORAGE GAS OF ZINC AND COPPER INDUSTRY

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STUDY OF THE THERMODYNAMIC PROPERTIES ABSORPTION SULFUR STORAGE GAS OF ZINC AND COPPER INDUSTRY

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***Abstract.** The article considers the problems of utilization of industrial gases formed in the production of copper and ore containing sulfur anhydrides. Currently, the composition of process gases entering the gas purification department is approximately 6-7% SO₂, 0.2-0.3% SO₃ and other gases, until sulfuric acid is obtained from these gases, the sulfate is treated in a counter-scheme with 8% sulfate acid. The aim is to first reduce the temperature of the exhaust gases and second to avoid flying components such as As, F, Se. As a result of the studies, the amount of SO₂ in the composition of the released process gases on the territory of the workshop was recognized as ineffective for the production of sulfuric acid. A number of scientific studies have been carried out on the production of other types of products, including the prevention of the release of harmful gases into the environment. At the same time, in order to fully utilize industrial gases containing sulfurous anhydrides, a method*

of absorption in a thin suspension was proposed, process kinetics and thermodynamic capabilities were studied. The results of thermodynamic calculations showed that among the chemical reactions taking place in the gas and solution systems, it was found that the reaction of sulfate acid for exposure to calcium carbonate was the slowest (limiting) stage for the general process and an optimal temperature of 60 ° C for the process was established.

Keywords: industrial gases, sulfur anhydrides, absorption, acute suspension, utilization, thermodynamics, mechanism, limiting stage, optimal temperature.

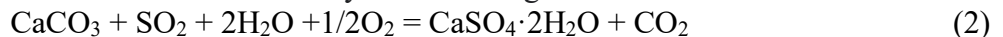
Introduction. In the process of production of zinc and copper metals in JSC "AMMC" emits a large amount of process gases containing sulfur dioxide [1]. The production of sulfuric acid from these gases is currently underway. Due to the non-hermetic nature of the exhaust gases in the metallurgical shop of the copper smelter of JSC "AMMC" and the fact that the process gases from the metallurgical units do not reach the department of complete degassing, it violates the environmental standards of the metallurgical plant. Today, the composition of process gases entering the gas purification department consists of about 6 - 7% SO₂, 0.2 - 0.3% SO₃ and other gases is given before the extraction of sulfuric acid from these gases is treated with 8% sulfuric acid according to the opposite scheme. The purpose is to reduce the temperature of the exhaust gases and to get rid of volatile components such as As, F, Se. As a result of the research, the amount of SO₂ in the process gases emitted in the area of the shop was found to be inefficient for the production of sulfuric acid. A number of scientific studies have been conducted on the production of other types of products in cases where the exhaust gases are low in sulfur dioxide, thereby preventing the release of harmful gases into the environment.

In many advanced industrial countries, including United States, Germany and Japan, other products containing low-land anhydride have been cleaned and manufactured [2-4].

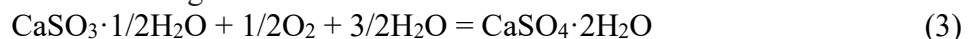
Based on the above information, it was proposed to recycle the process gases collected in the shop area using the absorption method. Accordingly, the process gases distributed throughout the metallurgical plant do not require excessive cooling. The process gases are sent directly to the gas treatment department [4-8]. The process gases containing purified sulfur dioxide interact with the lime in the absorber. In the absorber, the process gases containing sulfur dioxide, which move from bottom to top, interact with the limestone suspension supplied from above. As a result, sulfur (IV) oxide reacts with limestone to form calcium sulfite:



In addition, due to the presence of oxygen in the exhaust gases entering the absorber, a small amount of calcium sulfate is formed by the following reaction:



Exhaust gases from sulfur (IV) oxide are removed from excess moisture and released into the atmosphere through vertical pipes. Along with the reaction products, the limestone suspension collects in a circulating vessel at the bottom of the absorber. From here it is fed back to the sprinkler devices of the absorber. In this way, circulating irrigation is carried out with a limestone suspension. The rest of the plant is removed and sent to a gypsum system. Initially, sulfuric acid is added to oxidize the rest of the calcium carbonate in the bush and provide the desired pH. The total product is then fed to an oxidation tower with a compressed air supply system from the bottom. In this tower, the oxygen in the air and the calcium sulfites in the boot react with the following chemical reactions:



Materials and methods. The objects of study were industrial gases containing sulfur oxides and limestone suspension as an absorbent.

Microsoft Excel and Thermo Base were used to determine the thermodynamic values of chemical reactions in industrial gases and absorbent systems and to plot them in Ellingham.

Thermodynamic analyzes were performed taking into account the temperature dependence of the isobaric-isothermal potentials (Gibbs free energy). The change in Gibbs free energy (ΔG) is generally calculated using the following formula:

$$\Delta G_{\text{reak}} = \Delta H_{\text{reak}} - \Delta S_{\text{reak}} T \quad (\text{a})$$

Here: ΔH_{reak} – enthalpy of the corresponding chemical reaction, kJ/mol;

ΔS_{reak} – entropy of the corresponding chemical reaction, J/(mol·K);

T – is the absolute temperature of the system, K.

Depending on the Gibbs energy value of the system, the following conclusions can be drawn about whether the reactions proceed spontaneously under standard conditions and the direction of the reaction:

a) if the value of the change in Gibbs energy in a chemical reaction is $\Delta G_{298} > 0$, i.e. positive, these reactions cannot proceed spontaneously under standard conditions;

b) If $\Delta G_{298} < 0$, i.e. negative, these reactions proceed spontaneously under standard conditions. These reactions are economically and technologically feasible;

d) If $\Delta G_{298} = 0$, then the system is in a state of chemical equilibrium. You need to change the system parameters (P, T, C, V) to shift the balance in the direction of the desired reaction.

The temperature dependence of the equilibrium constant (K_M) of a reaction is determined by the following formula:

$$\Delta G = -RT \ln K_M \quad (\text{b})$$

Where: R is the universal gas constant, $R = 8.31696 \cdot 10^{-3}$ kJ/(grad · mol);

K_M is the equilibrium constant of the corresponding chemical reaction.

(b) To find the equilibrium constant from a mathematical expression, we logarithm both sides of the equation to RT, and the result is:

$$K_M = e^{-\frac{\Delta G}{RT}} \quad (\text{c})$$

Results and their discussion. Analysis of the mechanism of absorption of sulfur gases in limestone suspension.

According to the mechanism of absorption of gases at the liquid level, first the gases form a contact boundary with the liquid, then they begin to melt on the surface of the liquid, and finally the gas molecules diffuse along the volume of the liquid [8-13]. This distribution continues until an equilibrium state occurs in the system. Once the equilibrium is reached, the melting of the gases stops. According to Le Shatelier's principle, when a system in equilibrium is affected by an external factor, the equilibrium shifts in the direction that reduces that external influence. For example, solutions with an alkaline environment are used to ensure better absorption of sulfur-containing gases in liquids. This is because sulfur dioxide (SO_2) and sulfur trioxide (SO_3) react with the alkali in the solution due to their acidic environment and remain in the solution volume. This increases the solubility of the sulfur gases accumulated on the surface of the liquid in water, and the equilibrium shifts to the right (i.e., towards the melting of the gases).

Due to the high polarity of the sulfur trioxide molecule, its solubility in water and alkaline solutions is quite high. But the solubility of sulfur dioxide in water is relatively lower than the solubility of sulfur trioxide [14-15]. Therefore, the limiting stage (i.e., the slowest process) during the utilization of industrial gases containing sulfur in liquids by absorbing them in liquids is considered to be the dissolution of sulfur dioxide in water or other liquids. Solutions with different alkaline media can be used to increase the solubility of sulfur dioxide. However, not all of these solutions are cost-effective and therefore have not yet found their application. However, the annual increase in production capacity in many metallurgical enterprises requires the development of new and efficient technologies for the utilization of large amounts of toxic sulfur gases emitted into the atmosphere.

For the above reasons, a special technology for the efficient disposal of a mixture of sulfur-containing gases has been developed to address the shortcomings. This technology

ensures the absorption of sulfur gases in limestone suspensions in special containers around the production area.

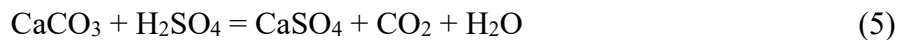
Since this process also has a limiting stage - the absorption of sulfur dioxide, this study focuses mainly on the thermodynamic aspects of this process.

(1), (2) and (3) In order to study the occurrence of chemical reactions in practice, it is first necessary to develop the most convenient and logically easy mechanism of this process. These chemical reactions generally occur in three stages:

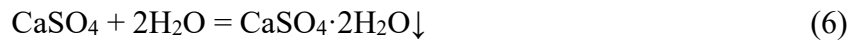
1. Sulfur dioxide, first dissolved in water, reacts with dissolved oxygen to form sulfuric acid:



2The sulfuric acid formed then reacts with the limestone particles in the suspension:



3. In the final stage, calcium sulfate salts bind water molecules and begin to precipitate as crystals (gypsum is formed):



This structured three-step mechanism can explain the process of chemical absorption of sulfur dioxide into a limestone suspension.

Thermodynamic analysis of chemical reactions in a system of sulfur gases and limestone suspensions

The thermodynamic values of all the substances involved in the reactions were calculated for standard conditions, and their initial values are given in Table 1.

Table 1.

Appropriate thermodynamic quantities of substances (at 298 K)

Substance	CaCO ₃	SO ₂	CaSO ₃	CO ₂
ΔH (kJ/mol)	-1206,8	-296,9	-929	-393,51
ΔG (kJ/mol)	-1128,4	-300,21	-903,7	-394,38
ΔS (J/(mol*K))	91,7	248,07	101,3	213,67
Substance	H ₂ SO ₄	H ₂ O	CaSO ₄	O ₂
ΔH (kJ/mol)	-911	-285,83	-1436,3	0
ΔG (kJ/mol)	-745,7	-237,25	-1323,9	0
ΔS (J/(mol*K))	18	70,08	106,7	205,04

The limiting stages in this case are the reaction of absorption of sulfur dioxide to the surface of the water (chemical equation 4) and the interaction of the resulting sulfuric acid with calcium carbonate (chemical equation 5). Because under normal conditions the equilibrium constant of the crystallization reaction (chemical equation 6) is greater than one, and under normal conditions the reaction proceeds spontaneously. Therefore, the study of the thermodynamic possibilities of the first two chemical reactions is sufficient to draw a general conclusion in the technological process.

The values of both chemical reactions for standard conditions were calculated according to Gess's law, and these values are given in Table 2.

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The values of both chemical reactions for standard conditions were calculated according to Gess's law, and these values are given in Table 2.

Table 2.
Calculated thermodynamic values of reactions 4 and 5 for standard conditions(at 298 K)

Thermodynamic quantities	Chemical equation 4	Chemical equation 5
$\Delta H_{\text{reak}} \text{ kJ}$	-328,27	2,16
$\Delta G_{\text{reak}} \text{ (kJ/mol)}$	-208,24	-81,43
$\Delta S_{\text{reak}} \text{ (J/K)}$	-402,67	280,75

From the values in Table 2, it can be seen that chemical reaction 4 is easier to proceed. Because the Gibbs energy value of the reaction is a negative number under standard conditions. The enthalpy of the reaction also has a negative value, ie the reaction produces heat (exothermic). The negative value of the entropy of the reaction indicates that the chaotic motion of the molecules slows down after the reaction.

Using these values, the general equation for the temperature-dependent Gibbs energy for chemical reaction 4 is:

$$\Delta G_T = -328,27 + 0,40267 \cdot T \quad (\text{d})$$

Table 3.
Changes in the thermodynamic values of chemical reaction 4 in the temperature range

№	ΔH	R	ΔS	T	ΔG_T	$\text{Ln}K_M$	K_M
1	-328,3	8,31	-0,403	300	-207,5	0,0832	1,0868
2	-328,3	8,31	-0,403	310	-203,4	0,079	1,0822
3	-328,3	8,31	-0,403	320	-199,4	0,075	1,0779
4	-328,3	8,31	-0,403	330	-195,4	0,0713	1,0738
5	-328,3	8,31	-0,403	340	-191,4	0,0677	1,0701
6	-328,3	8,31	-0,403	350	-187,3	0,0644	1,0665
7	-328,3	8,31	-0,403	360	-183,3	0,0613	1,0632
8	-328,3	8,31	-0,403	370	-179,3	0,0583	1,06
9	-328,3	8,31	-0,403	380	-175,3	0,0555	1,0571
10	-328,3	8,31	-0,403	390	-171,2	0,0528	1,0543
11	-328,3	8,31	-0,403	400	-167,2	0,0503	1,0516
12	-328,3	8,31	-0,403	410	-163,2	0,0479	1,0491
13	-328,3	8,31	-0,403	420	-159,1	0,0456	1,0467
14	-328,3	8,31	-0,403	430	-155,1	0,0434	1,0444
15	-328,3	8,31	-0,403	440	-151,1	0,0413	1,0422

Table 3 shows the change in the free energy value of the reaction in the temperature range of 300–440 K (or 27–167 °C) for chemical reaction 4, and, at the same time, the change in the equilibrium constants of the reaction. It was calculated that since the 4th chemical reaction was exothermic, the increase in temperature had a negative effect on the rate of the chemical reaction.

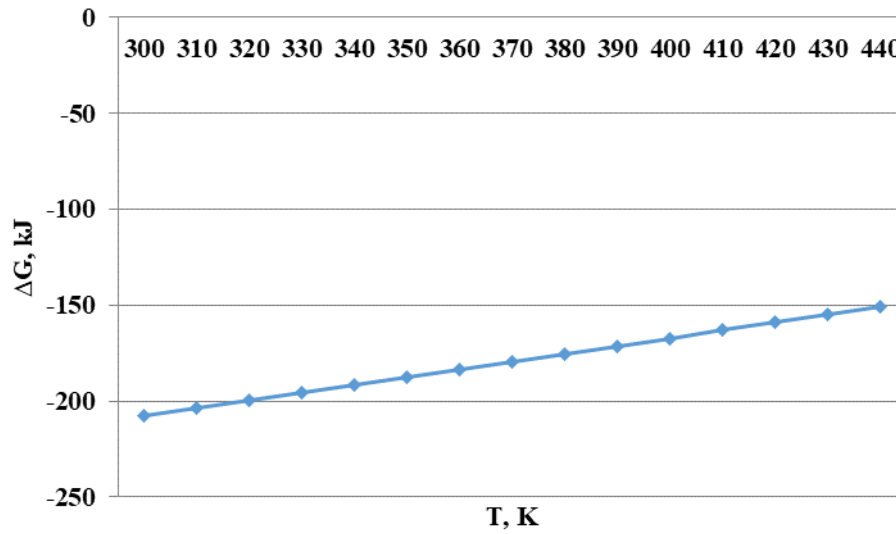


Figure 1. Temperature dependence diagram of Gibbs energy for reaction 4

From the graph shown in Figure 1, it can be seen that increasing the temperature in the reaction system by every 10 units led to a positive Gibbs energy of the reaction. In this graph, chemical reaction 4 at a temperature of 300 K (or 27 °C) has a minimum value of Gibbs energy, and this temperature provides the most favorable opportunity for the process to flow. As a proof of this, it can be seen in Figure 2 that the equilibrium constant of a chemical reaction at a temperature of 300 K has a maximum value (1.0868).

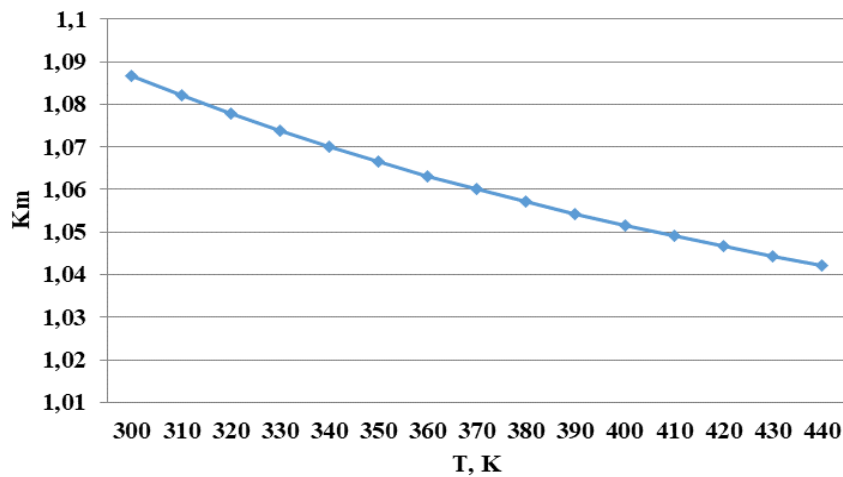


Figure 2. Temperature dependence diagram of the equilibrium constant for reaction 5

The enthalpy and entropy values of chemical reaction 5 are positive, which means that the initial activation energy is required for it to flow spontaneously under standard conditions. Therefore, in this case, chemical reaction 5 is a limiting step for the process that takes place in the kinetic mode, and it is important to study the thermodynamic possibilities of this reaction.

Using the values given in Table 2, the general Gibbs temperature-dependent equation for chemical reaction 5 was constructed:

$$\Delta G^T = 2,16 - 0,28075 \cdot T \quad (e)$$

According to the given mathematical expression, the possibility of chemical reaction 5 occurring at several temperatures has been determined.

Table 4.

Changes in thermodynamic values in the temperature range

N ₂	ΔH	R	ΔS	T	ΔG_T	$\ln K_M$	K_M
1	2,16	8,31	0,2808	300	-82,07	0,0329	1,0335
2	2,16	8,31	0,2808	310	-84,87	0,0329	1,0335
3	2,16	8,31	0,2808	320	-87,68	0,033	1,0335
4	2,16	8,31	0,2808	330	-90,49	0,033	1,0335
5	2,16	8,31	0,2808	340	-93,3	0,033	1,0336
6	2,16	8,31	0,2808	350	-96,1	0,033	1,0336
7	2,16	8,31	0,2808	360	-98,91	0,0331	1,0336
8	2,16	8,31	0,2808	370	-101,7	0,0331	1,0336
9	2,16	8,31	0,2808	380	-104,5	0,0331	1,0337
10	2,16	8,31	0,2808	390	-107,3	0,0331	1,0337
11	2,16	8,31	0,2808	400	-110,1	0,0331	1,0337
12	2,16	8,31	0,2808	410	-112,9	0,0332	1,0337
13	2,16	8,31	0,2808	420	-115,8	0,0332	1,0337
14	2,16	8,31	0,2808	430	-118,6	0,0332	1,0337
15	2,16	8,31	0,2808	440	-121,4	0,0332	1,0338

The changes in the free energy and equilibrium constants of the reaction of sulfuric acid solution with calcium carbonate at several temperatures are presented in Table 4. In this case, the values of Gibbs energy and equilibrium constant of the reaction are calculated when the temperature in the system changes every 10 units.

The results of the thermodynamic analysis presented in Table 4 show that since the reaction of a sulfuric acid solution with calcium carbonate is an endothermic reaction, an increase in temperature in the system increases the rate of the chemical reaction going in the right direction. increases according to the law.

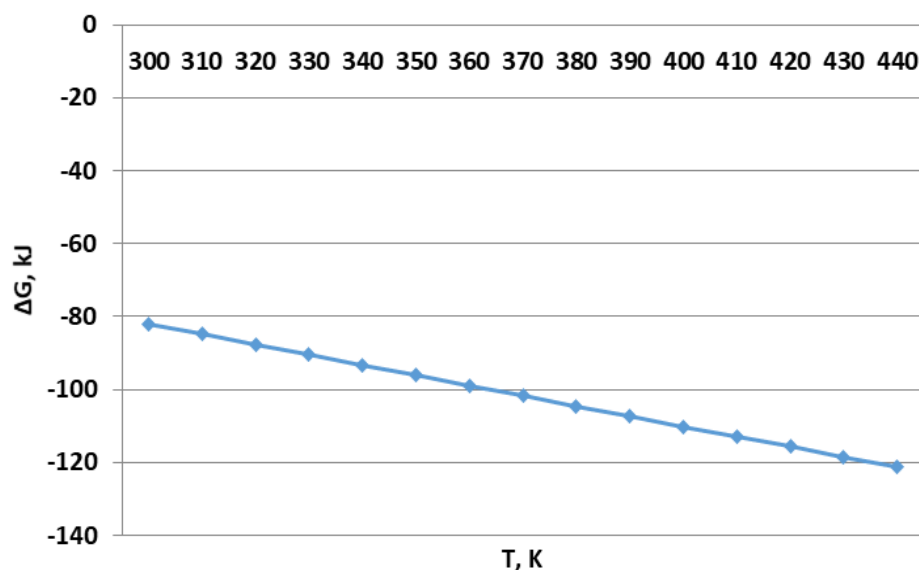


Figure 3. Temperature dependence diagram of Gibbs energy for reaction 5

The value of the Gibbs energy in a system is a thermodynamic parameter that determines the probability of a reaction occurring or not at a given temperature. The graph in Figure 3 illustrates the temperature dependence of the Gibbs energy for reaction 5, and it can be seen from this graph that the free energy (ΔG) of the system for this condition decreases linearly when the temperature changes every 10 units, i.e. becomes negative. This means that as the temperature rises, the reaction of sulfuric acid with calcium carbonate increases the likelihood of leakage.

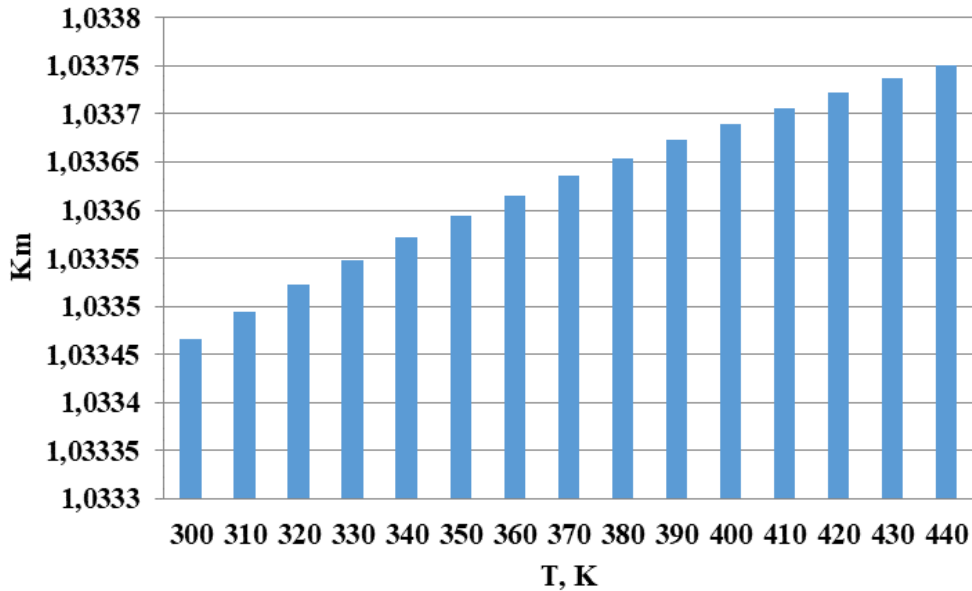


Figure 4. Temperature dependence diagram of the equilibrium constant for reaction 5

Figure 4 shows a graph of the temperature dependence of the equilibrium constant of reaction 5. This graph shows an increase in the value of the chemical equilibrium constant between 300 and 440 K, and a stabilization of the curve at temperatures above 380 K. It should be noted that the value of the chemical equilibrium constant at each given temperature contains values that do not differ much from one another, i.e., the rate of the chemical reaction is not that high. Therefore, the reaction of sulfuric acid with calcium carbonate is the main limiting step that determines the speed of the process. In this case, the acceleration of the reaction depends on the number of collisions between the molecules of the reactants. This is determined by the rate of diffusion of substances and the size of the reaction surface.

Using a thermodynamic analysis of all the above reactions, an Ellingham diagram was constructed for all the chemical reactions that take place during the absorption and utilization of sulfur dioxide in a limestone suspension, as shown in Figure 5.

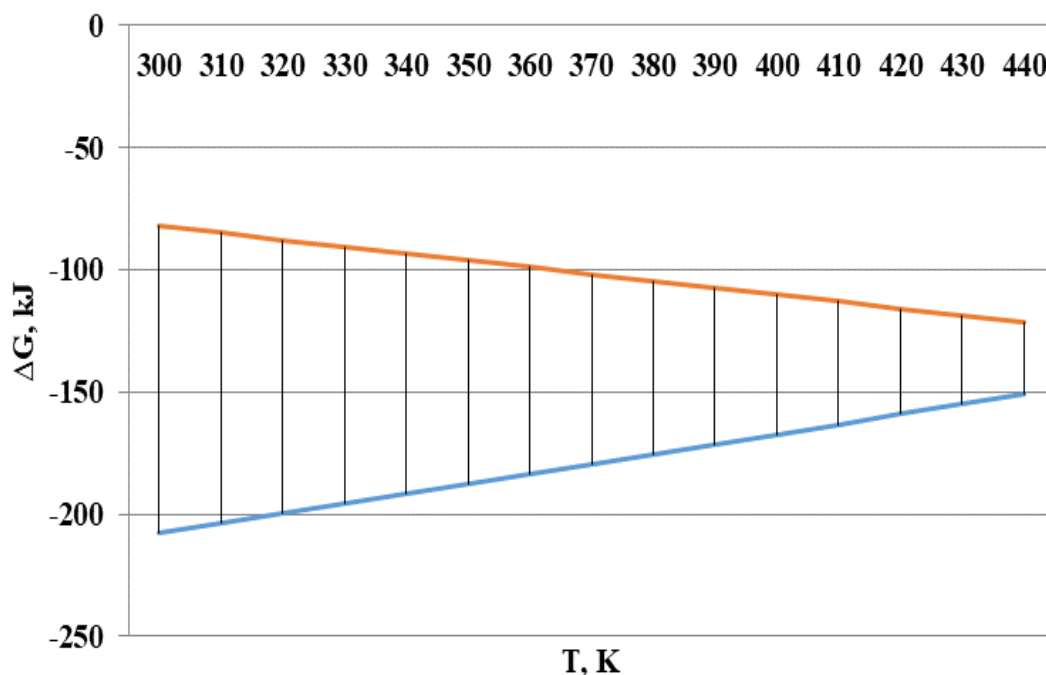


Figure 5. General Ellingham diagram of chemical reactions in a gas-solution system

Since the slowest (limiting) reaction of all the chemical reactions studied in a sulfur gas and limestone suspension system is the reaction of sulfuric acid with calcium carbonate, the optimal temperature of the system is determined by this reaction temperature. From the graph shown in Figure 5, we can see that, given the endothermic reactions, the probability of a faster reaction of sulfuric acid with calcium carbonate corresponds to a temperature of 60 °C (333 K). At this temperature, other chemical reactions are more likely to occur.

Conclusion. Absorption technology for the utilization of toxic effluents generated during the pyrometallurgical production of non-ferrous metals has been developed. Accordingly, sulfur dioxide and sulfur trioxide emissions from industrial gases have been found to cause significant damage to the environment.

The use of a limestone suspension as an economically inexpensive absorbent for the efficient utilization of industrial gases containing sulfur oxides has been studied.

An easy and convenient logical mechanism for the chemical absorption of sulfur dioxide in a limestone suspension has been developed.

According to the developed mechanism, thermodynamic aspects of chemical reactions flowing in gas and solution systems were studied. Accordingly, the reaction of sulfuric acid with calcium carbonate was found to be the slowest (limiting) stage for the overall process, and an optimal temperature of 60 °C (333 K) was set for the process.

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SYSTEM THINKING IN ANALYSIS OF DEVICE FOR REMOVING MOISTURE FROM THE MATERIAL BY INSTANTANEOUS PRESSURE RELEASE

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Abstract. The article covers the solution of the problems of systematic analysis and mathematical modeling of the drying process based on increasing and instantaneous lowering the pressure in the dryer. The drying process and devices are analyzed based on systematic thinking, and the systematic analysis of the technology of drying of agricultural products is studied in terms of quasi-apparatus and quasi-layers of the product.