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CONTACT OXIDATION OF CONCENTRATED SO_2 MIXTURES – I

The paper is concerned with an investigation of the catalytic oxidation sulfur dioxide in a multi–layer adiabatic reactor included in a cyclic circuit. The oxidation of sulfur dioxide by oxygen in the contact mass layer was investigated by computational analysis of a mathematical process description. In part – I of the study the effect of the composition of the reactive mixture on the efficiency of the oxidation process was investigated. Also the composition resulting in the maximum and specific capacity, as well as the minimum contact mass volume of the layer was determined.

The technology of the sulfuric acid production by the contact method is widely applied and has been extensively studied [1–6]. The most important stage of this method is the process of sulfur dioxide oxidation, usually performed on the vanadium catalysts in a reactor with adiabatic layers of the contact mass. The reactive gas (usually composed in % vol. of SO_2 : 7–9, O_2 : 9–14, N_2 : 79–82), used for making contact passes in succession through 3 – 5 layers of the contact mass which is cooled by heat exchangers situated between the layers. The obtained contact gas passes to the stage of SO_3 release and then the exhaust gas is released into the atmosphere after being cleaned of SO_2. In essence this system has become classical.

It is known that the main amount of SO_3 (about 70%) in the classical system is obtained at the first layer of the catalyst [4]. The rate of SO_2 oxidation at the subsequent layers is much lower than in the first layer. Therefore the greater part of the contact mass is used unproductively but is never the less needed for attainment of the required full conversion of SO_2. The final conversion of SO_2 into SO_3 is the most important performance index of the contact oxidation process. As this takes place, the efficiency of the contact separation and the contact mass volume are put in dependence on the specified index [1, 4, 7]. The strict requirements applied to the final SO_2 conversion essentially limit the concentration ranges of SO_2 and O_2 at the inlet into the layer in the classical system; the SO_2 concentration in the reactive gas at the inlet into the layer cannot be higher than 12% vol, and the O_2 concentration not higher than 15% vol.

The possibility for processing reactive gases with high contents of SO_2 and O_2 by the contact method has been described [1, 2, 5, 6]. However, either the application of heat resistant catalysts and increased pressure [12, 13] or the isolation of the technological system concerning exhaust gas [8, 14–19] are required. Besides, the high intensity of heat release during the oxidation of concentrated mixtures complicates the heat control of the layers and isolating the system is connected with additional expenses for the reycling line and the air separation plant.

It is possible to solve the problems of excessive heat evolution and the additional expenses. Cyclic circuits [15, 16] in which the concentrated sulfide mixture subsequently passes through 3 – 5 layers of the contact mass, all operating in the same temperature mode, are well known. The capacity of the contact plant substantially increases from layer to layer. However, as a consequence of SO_3 concentration buildup in the contact gas from layer to layer, the SO_2 oxidation rate drops resulting in a drastic drop of the specific capacity of the contact mass.

At the same time, there is no need to maintain a high SO_2 fractional conversion in the circuits with exhaust gas recirculation. In [15] the authors centered on a value of X equal to 60%. The limiting requirements of the final conversion in the processing of concentrated sulfide mixtures in a cyclic circuit opens the way for increasing the capacity of the contact plant and for reducing the required contact mass volume of each layer at the cost of the correct choice of the optimum gas load for the plant and the even distribution of the reactive load along the layers. As a result, the specific capacity of the contact mass in the layers increases, and the plant dimensions and it pressure loss decrease, which are important constituents to compensate for the expenses, incurred to install the air separation plant and to arrange the recyding.

Increasing the ranges of the sulfur dioxide and oxygen concentrations, as well as the sulfur trioxide availability in the reactive gas at the inlet to the layer extend the choice of the controlling parameters. Control the heat mode of the layer effectively and reducing the

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heat release can be realised. This offers considerable scope for setting up technological circuits for SO_2 contact oxidation using the concentrated sulfide mixtures as the reactive gas. Of special note is the increasing ecological safety of cyclic circuits. Recycling the exhaust contact gas for processing enables for the amount of SO_2 released to the atmosphere to be substantially reduced and high ecological results to be achieved [14]. It is becoming possible to consider cyclic circuits as being of considerable promise for introduction in industry.

The present article is devoted to the analysis of the oxidation of concentrated sulfide mixtures in a contact plant operating exhaust gas recycling. The purpose of the study was to evaluate the performance of concentrated sulfide mixture processing and the practical possibility of the implementation of cyclic technology.

In the article:

- The oxidation of various concentrated sulfide mixtures in the adiabatic layer of the contact mass was investigated by means of computational analysis. The result of the computations are compared with the oxidation of the gas – air mixture of composition (% vol.): $SO_2=9$, $O_2=12$, $N_2=79$ used in the classical circuit.
- The effect of the reactive gas consumption upon the value of the oxidation performance of the concentrated sulfide mixtures in the layer of the contact mass was analysed. The composition of the mixture, resulting in the best value of the capacity, specific capacity and volume of the contact mass layer were determined.
- The controllability of the contact oxidation of concentrated sulfide mixtures was studied. The limits of changing the composition and the reactive gas consumption were found from the condition of providing the specified temperature mode.
- Schematic flow systems of the contact oxidation of concentrated sulfur dioxide have been suggested in which high results and effective control can be achieved.

MATHEMATICAL DESCRIPTION OF THE PROCESS

A study of the oxidation process of the concentrated sulfide mixtures in the contact mass adiabatic layer has been performed by the use of the mathematical model constructed on the basis of regularities and described in [9, 10]. In constructing the mathematical description of the oxidation process in the contact mass layer it was suggested that heat exchange process between the gas phase and the contact mass surface should be taken as ideal, the contact mass as homogeneous and the oxidation as kinetically controlled. The following is accepted: the catalyst activity, the viscosity and density of the flowing gas, the homogeneity of the temperature profile at the inlet and the radial profile of the rates in a section of the layer are all considered to remain constant. The chemical reaction occuring on the contact mass surface is:

$$2 \cdot SO_2 + O_2 = 2 \cdot SO_3 \tag{1}$$

Within the accepted limits, the oxidation process ($SO_2 \rightarrow SO_3$) in the catalyst layer is characterized by the following equations:

$$\frac{dN_i}{dl} = \nu \cdot r \cdot S \tag{2}$$

$$\frac{dT}{dl} = \frac{(Q_p(T^0) - C^* \cdot (T - T^0) \cdot \nu) \cdot r \cdot S}{N \cdot C(T)}; \tag{3}$$

$$\nu = \sum_{i=1}^{3} \nu_i,\ N = \sum_{i=1}^{3} N_i,$$

$$C(T) = \sum_{i=1}^{3} N_i \cdot C_i(T) / N, \quad i \in \{SO_2,\ O_2,\ SO_3\}$$

The rate of the chemical reaction r(1), expressed through the partial pressures of the reactant, is defined by the formula [3]:

$$r = K \cdot \frac{P_{O_2}}{1 + 0.8 \cdot P_{SO_3}/P_{SO_2}} \left(1 - \left(\frac{P_{SO_3}}{P_{SO_2} \cdot P_{O_2}^{0.5} \cdot K_e} \right)^2 \right) \tag{4}$$

A characteristic feature of SO_2 contact oxidation in a cyclic circuit is that not the final conversion of SO_2 in SO_3 is related to the capacity, specific capacity and volume of the CM (contact mass) layer as in [11] but the process temperature mode. In this case the following conditions are imposed on the temperature mode:

$$T^f - T^0 + \Delta = T_d - T_i; \tag{5}$$

$$T_i \le T^0 < T^f \le T_d; \tag{6}$$

$$0 < \Delta \le \Delta^p, \tag{7}$$

These result from the following circumstances. The temperature T^0 of the mixture at the layer inlet cannot be below the ignition temperature T_i of the reactive mixture at the catalyst. At the same time, the temperature T^f at the outlet from the layer should not exceed the threshold value of T_d, above which thermal decomposition of the contact mass begins. Condition (5) defines the agreement of the temperature drop (T^f–T^0) at the layer with the allowable drop (T_d–T_i). Here the value Δ determines the reserve for overheating and cooling the contact mass. It cannot be lower than zero. Simultaneously, the indices of the process are dependent on the specified value. So, the capacity of the layer Ω, according to the heat balance of the SO_2 oxidation process, is connected with the temperature drop (T^f–T^0) as follows:

$$\Omega = \int_{T^0}^{T^f} \frac{N \cdot C(T) \cdot dT}{Q_r(T^o) - C^* \cdot (T - T^o) \cdot \nu}. \tag{8}$$

In their turn, the contact mass volume of the layer and its specific output depend on the value Ω:

$$V = \int_{N'^{o}_{SO_3}}^{N'^{o}_{SO_3}+\Omega} dN_{SO_3}/W \qquad (9)$$

$$\omega = \Omega/V. \qquad (10)$$

From (8) it follows that the lower is T^0 and higher is T^f, the greater the integrand and, consequently, the value Ω. Thus, the maximum capacity of the contact mass adiabatic layer is given by the temperature mode wherein $T^f = T_d$ and $T^0 = T_i$ and the lower is Ω, the greater the layer capacity. However, SO_2 oxidation in the temperature mode at $\Delta=0$ is dangerous, on the one hand, owing to the possibility of catalyst overheating and on the other hand, owing to possibility of feeding cool gas to the inlet into the layer and stopping the SO_2 oxidation reaction within the contact mass. The temperature mode of the oxidation process providing the capacity of the contact mass layer approaching the maximum value, avoiding catalyst overheating and cooling is determined from (5) by setting the value $\Delta \leq \Delta^p$.

ANALYSIS OF THE OXIDATION PROCESS OF THE CONCENTRATED MIXTURES

A study of the SO_2 oxidation process in the catalyst layer was performed for the given initial data: gas temperature at the inlet to the layer was T^0=723K, at the outlet T^f=873K, layer diameter – 6 m, gas consumption – 100 mol/s. Computation of the operating mode of the layer was performed for the following sulfide mixtures (Table 1): mixture 1 was obtained by burning sulfur in air; mixtures 2 – 13, were concentrated mixtures of SO_2 and O_2 with different ratios of these components; mixtures 14–17, in addition to SO_2 and O_2, also contained SO_3.

The obtained results were first and foremost considered in the context of the possibility of realizing the concentrated gas oxidation process in a temperature mode defined by conditions (5)–(7). It is well known [3] that while processing gas–air mixtures the true contact time τ^{tr} of a reactive gas with the catalyst surface exceeds the theoretical time τ^{th} of the contact which is needed to achieve a preset degree of conversion. At the expense of the specified condition, the finite degree of SO_2 fractional conversion at the outlet from the layer is close to the equilibrium value. A comparison of the finite and equilibrium value of the fractional conversions makes it possible to judge a condition of the oxidation process in processing concentrated sulfide mixtures.

Similar to the gas–air mixture 1, mixtures 2, 13–17 at outlet from the layer have a finite functional conversion at the outlet close to the equilibrium value. The processing of such concentrated mixtures within the limit of (5)–(7) does not cause problems as the value of Δ in these cases cannot be negative and satisfies condition (7). Notice, that it is possible to gain execution of condition (7), as well as the proximity of the values of the finite and equilibrium fractional conversions at the outlet from the layer, by either limiting concentration of one of the reagents at the inlet to the layer (mixtures 2, 13) or at the expense of SO_3 being present in the reactive mixture (mixtures 14–17), i.e. by shifting the reaction equilibrium (1) to it decomposition side.

Table 1. Specifications of sulfuric mixture oxidation process in the contact mass adiabatic layer (N=100 mole/s, T^0 = 723 K, T^f = 873 K)

Srl No	Mixture composition, % vol.				Fractional conversion		V	Ω	w
	SO_2	O_2	SO_3	N_2	X^o	X^f	m^3	mol/s	mol/ /(s·m^3)
1	9.0	12.0	0.0	79.0	0.724	0.737	24.36	6.721	0.290
2	9.24	90.8	0.0	0.0	0.886	0.899	4.125	8.310	2.014
3	13.0	87.0	0.0	0.0	0.650	0.897	2.713	8.640	3.147
4	15.0	85.0	0.0	0.0	0.560	0.896	2.685	8.660	3.223
5	16.6	83.4	0.0	0.0	0.510	0.895	2.713	8.760	3.229
6	19.0	81.0	0.0	0.0	0.450	0.893	2.769	8.820	3.185
7	23.0	77.0	0.0	0.0	0.380	0.890	2.854	9.000	3.153
8	43.0	57.0	0.0	0.0	0.220	0.865	4.041	9.650	2.388
9	53.0	47.0	0.0	0.0	0.180	0.841	5.030	9.860	1.965
10	63.0	37.0	0.0	0.0	0.160	0.790	6.585	10.019	1.522
11	73.0	27.0	0.0	0.0	0.140	0.655	9.608	10.750	1.119
12	85.5	14.2	0.0	0.0	0.120	0.332	19.50	10.760	0.552
13	94.6	5.4	0.0	0.0	0.113	0.114	110.2	10.750	0.098
14	14.4	65.6	20.0	0.0	0.879	0.882	13.85	10.340	0.747
15	16.9	55.1	28.0	0.0	0.867	0.870	19.49	11.150	0.572
16	17.7	52.3	30.0	0.0	0.865	0.867	21.19	11.400	0.538
17	37.6	12.8	49.6	0.0	0.721	0.781	89.02	13.400	0.151

The processing of mixtures 3–12 occurs away from equilibrium. For example, for a mixture composed of (% vol.) SO_2 = 43, O_2 = 57, the finite fractional conversion of SO_2 within the limits of the temperature mode restricted by conditions (5)–(7) is 0.22 whereas the equilibrium fractional conversion at a temperature of 873K is 0.865. Intensive oxidation of SO_2 occurs in such mixtures. In actual practice, when processing such mixtures, the value of Δ may become negative resulting in a great danger of catalyst overheating because of the existence of local temperature gradients in the cross–section of the layer and the concentration of the reagents due to heterogeneity of, for example, contact mass, gas flow density.

To avoid the danger of catalyst overheating when processing mixtures like 3–12, it is necessary to maintain value the of τ^{tr} (the real contact time of the reactive gas with the catalyst) defined by the reactive gas consumption at a value that is not higher than the value of τ^{th} (the theoretical contact time). In other words, the condition $\Delta > 0$ has to be fulfilled. Since the theoretical contact time is defined by the reactive gas composition and selected according to conditions (5)–(7), then it is practicable to perform the processing of the concentrated mixture without overheating the catalyst by acting upon τ^r by changing the reactive mixture consumption and upon τ^{th} by changing the mixture composition.

Now compare the data given in Table 1 in the context of the efficiency of the processing of various mixture. It can be seen that the indices of Ω, V and ω, whereby the oxidation of concentrated sulfide mixtures can be judged adequately, differ substantially not only from the corresponding indices of gas–air mixture processing, but also between them. Furthermore, the nature of the dependence of the efficiency indices on the gas composition at the inlet to the contact layer is different. So the capacity increases monotonically with increasing content of sulfur oxides in the reactive mixture whereas the contact mass volume has a minimum and, on the contrary the specific capacity has a maximum.

The influence of the reactive mass composition on the values indices of SO_2 oxidation process efficiency and aspects of the realization of the oxidation of the concentrated sulfide mixtures considered above are central to the the problems of control over the oxidation process and of structural organization of the contact section. In this connection, a study of the mechanism of the influence of the reactive gas composition on the capacity, volume and specific capacity of the contact mass layer is necessary.

CAPACITY OF LAYER

An essential distinction between concentrated mixture processing and air oxidation of sulfur dioxide is the dependence of the layer capacity on the SO_2:O_2 relationship and the SO_3 content in the reactive gas at the inlet into the contact layer. The lowest capacity of the layer is when processing SO_2 in a mixture with air. The processing of a mixture of content (% vol.): SO_2=9.24, O_2=90.76 increases the capacity 1.2 times; of content: SO_2=94.6, O_2=5.4 – 1.6 times: of content: SO_2=37.6, O_2=12.8, SO_3=49.6 – 2 times.

The influence of the SO_2 and SO_3 concentrations in the original gas on the layer capacity is shown by the data in Table 2 and 3 and in Figures 1 and 2. As illustrated in Fig. 2, dependence of the capacity of the layer on the SO_3 content in the reactive mixture has an extreme form. This is explained by the influence of the gas mixture heat capacity and reaction reversibility (1) on Ω. Let us consider it.

With increasing SO_3 content in the gas, its heat capacity increases enabling the oxidation of a greater amount of SO_2 in the adiabatic layer. As compared to gas – air mixtures, a substantial variation of the heat capacity of the concentrated mixtures processed is possible. The heat capacity of gas of content (% vol.): SO_2=8.26, O_2=15, N_2=76.74 at 500°C is equal to 3387 J/(K · mol), whereas the heat capacity of the concentrated mixture: SO_2=8.26, O_2=91.74 is 3710 J/(K · mol); for mixture: SO_2=43, O_2=57 – 4400 J/(K · mol) and in case of the mixture SO_2=94.64, O_2=5.36 – 5000 J/(K · mol). The difference between the heat capacities is almost 1.5 times. With increasing temperature within the range 450 – 600°C, the increase in the heat capacity of the components of the mixture will be: SO_2=4.01, O_2=1.5 and SO_3=8.53 J/(K · mol).

The increase in the layer capacity is limited by the equilibrium of the reversible chemical reaction (1). As

Table 2. Effect of SO_3 concentration in the reactive mixture on the layer performance efficiency (N=100 mol/s, SO_2:O_2 = 1:1, T^o = 723 K, T^f = 873 K)

No	Mixture composition, % vol.			V	ω	Ω
	SO_2	O_2	SO_3	m^3	mol/ /(s · m^3)	mol/s
1	50.0	50.0	0.0	4.66	2.058	9.595
2	45.0	45.0	10.0	6.98	1.525	10.642
3	40.0	40.0	20.0	9.95	1.140	11.343
4	35.0	35.0	30.0	15.09	0.791	11.941
5	30.0	30.0	40.0	24.64	0.519	12.794
6	25.0	25.0	50.0	56.24	0.222	12.489
7	20.0	20.0	60.0	93.26	0.109	10.180
8	15.0	15.0	70.0	178.04	0.043	7.580
9	10.0	10.0	80.0	452.16	0.01	4.540

Table 3. Influence of SO_2:O_2 relation in the reactive mixture on the layer performance efficiency (N = 100 mol/s, T^o = 723 K, T^f = 873 K)

No	Mixture composition, % vol.		V	ω	W
	SO_2	O_2	m^3	mole/(s · m^3)	mole/s
1	10.0	90.0	2.911	2.891	8.410
2	20.0	80.0	2.797	3.168	8.860
3	30.0	70.0	3.193	2.864	9.146
4	40.0	60.0	3.815	2.516	9.600
5	50.0	50.0	4.691	2.093	9.820
6	60.0	40.0	6.048	1.665	10.070
7	70.0	30.0	8.365	1.225	10.250
8	80.0	20.0	13.226	0.797	10.540
9	90.0	10.0	29.810	0.361	10.750

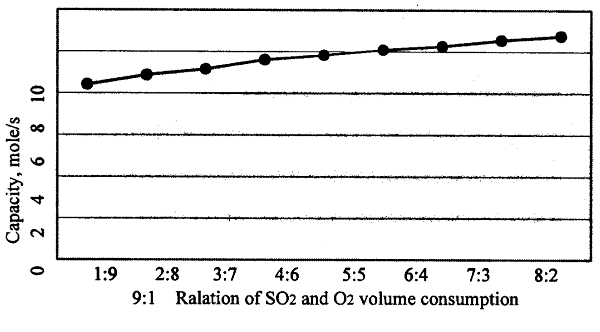


Figure 1. Dependence of the capacity on the SO_2:O_2 relation in the reactive gas with 100 mol/s consumption without SO_3

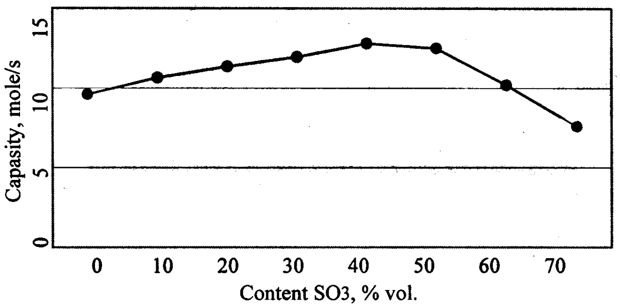


Figure 2. Dependence of the capacity on the SO_3 content in the reactive gas with 100 mol/s consumption, SO_2:O_2=1:1

SO_3 is accumulated in the contact gas, the growth of Ω is decreased and finally ceases when the gas in the layer reaches the equilibrium composition. The equilibrium of the reversible chemical reaction (1) is characterized by the equilibrium constant $K_e = P^e_{SO_3}/(P^e_{SO_2}\sqrt{P^e_{O_2}})$. The result of the oxidation is defined by the equilibrium fractional conversion of SO_2 in SO_3:

$$X^e = K_e/(K_e + 1/\sqrt{P^e_{O_2}}) \tag{11}$$

$P^e_{O_2}$ can be expressed in terms of Ω: $P^e_{O_2} = P \cdot (N^e_{O_2} - 0.5 \cdot \Omega^e)/(N^0 - 0.5 \cdot \Omega^e)$. Then substituting $P^e_{O_2}$ in (11) and taking into account that $X^e = (N^0_{SO_3} + \Omega^e)/(N^0_{SO_2} + N^0_{SO_3})$ the desired dependence; after rearrangement, is obtained:

$$\Omega^e = \frac{K_e(N^0_{SO_2} + N^0_{SO_3})}{K_e + 1/\sqrt{[(N^0_{O_2} - 0.5 \cdot \Omega^e)/(N^0 - 0.5 \cdot \Omega^e)]}} - N^0_{SO_3}. \tag{12}$$

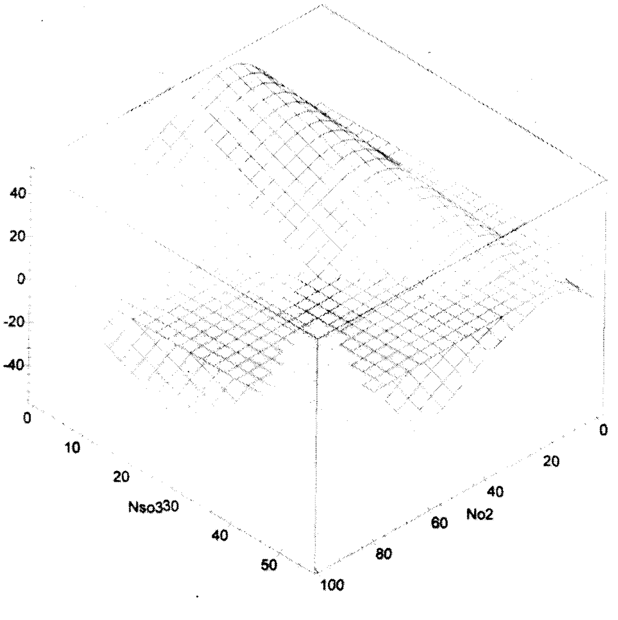


Figure 3. Effect of the reactive gas composition on the capacity

Letting N = const and excluding N_{SO_2} from (8) and (12) by means of substituting $N_{SO_2} = N - N_{O_2} - N_{SO_3}$, it is possible to construct Ω in three-dimensional space as a function of the reactive mixture composition at the input to the layer. The result is shown in Fig.3. Here, equation (8) has the shape of an inclined plane and (12) an implicit relation of a convex non-linear surface. At the intersection of the plane and surface, a line is formed on which Ω reaches its maximum value.

During processing the reactive mixtures, when the sulfur dioxide conversion stage obtainable in the layer is less than the equilibrium one, the increase in SO_2 and SO_3 in the reactive gas causes Ω to increase under the effect of the gas heat capacity. As the actual stage of sulfur dioxide conversion in the layer approaches the equilibrium value, the increase in Ω ceases. This condition is illustrated in Fig. 3 from which it can be seen that the increase in Ω ceases on the line of the intersection of the plane and the non-linear surface. On passing over the intersection line of the plane and the non-linear surface, the actual stage of sulfur dioxide conversion cannot exceed the equilibrium one and Ω begins to decrease. This occurs either due to an increase in SO_3 in the gas or due to a lack of SO_2 or O_2 in the reactive mixture. Thus, the influence of the factors considered determines the extreme value of the capacity. Computations show that the maximum capacity of the layer is given by a reactive gas of content (% vol.): SO_2=37.6, O_2=12.8, SO_3=49.6. Thus, 13.4 mol/s of SO_3 is produced in 89 m^3 of the contact mass when processing 100 mol/s of such gas. As this takes place, the specific capacity of this layer is 0.15 mol/(s · m^3), which is 1.93 times less than that of the first layer when processing a gas-air mixture in an open circuit. However, on oxidation of SO_2 in a cycle circuit with one layer whose contact mass volume is equal to the volume of four layers in the contact plant of an open circuit, it is possible to obtain twice as much SO_3. Thus the specific capacity of the layer contact mass when processing concentrated sulfurous gas of composition providing the maximum capacity is higher than the specific capacity when processing mixture 1 in a classical circuit.

THE CONTACT MASS VOLUME AND SPECIFIC CAPACITY

Concerning the contact mass volume, the most effective is the processing of the concentrated mixtures with an excess of oxygen (ref. to Table 1). Thus, to obtain 8.31 mol/s of sulfur trioxide from 100 mol/s of a reactive mixture containing (% vol.): SO_2=9.24, O_2=90.76, the contact mass required is 5.9 times less than that needed to obtain 6.72 mol/s of sulfur trioxide from the gas-air mixture.

The contact mass volume of the layer is determined according to (9) by W, the rate of chemical reaction (1), and the capacity required. The value W is connected with the parameters of the process in an intricate manner: through the gas content and temperature. The nature of this connection is described in (4). The temperature of the process, which varies from the top to the bottom of the layer, affects the result through the velocity constants and the equilibrium of reaction (1). The equilibrium (2) and (3) determine the relationship between the change in temperature along the height of the contact mass layer and the change in the reactive gas composition. In the end, the desired value of V is presented by a function, which depends on the reactive gas consumption, its original composition and the temperature of the process. The contact mass volume was found by numerical methods, given in [9], specifying the gas consumption, its composition and the temperature at the layer input and performing the process according to (5)–(7).

The dependence of the contact mass volume on the SO_3 content in the reactive mixture at the layer input is shown in Fig. 4 (Table 2). The reactive gas consumption through the layer was 100 mol/s and the SO_2:O_2 ration was 1:1. The dependence of the contact mass volume required on the SO_2:O_2 ratio for a reactive mixture free from SO_3 is shown in Fig. 5 (Table 3). The presented dependence has a flattened minimum when, according to the calculations, the reactive mixture has the composition (% vol.) SO_2=15, O_2=85. As this occurs 8.66 mol/s sulfur trioxide is produced in a layer of 2.72 m^3 volume.

The specific capacity of the layer defines the amount of SO_3 obtained in the residence time of the reactive gas in a unit volume of the contact mass of the layer. Analysis of the calculated data testifies that the required contact mass volume increases faster with increasing SO_3 content than the growth of the layer capacity. Therefore, the specific capacity is more effected by changes in V than by changes in the capacity.

Fig. 6 (Table 2) shows the dependence of the specific capacity on the available SO_3 in the reactive gas and Fig. 2 shows the change of the layer capacity complying with it. As is the case with the contact mass volume, the absence of SO_3 in the reactive gas at the layer input has a beneficial effect on ω (the layer specific capacity). The influence of the SO_2:O_2 on ω ratio is shown in fig. 7 (Table 3). The value of ω attains a maximum with a reactive gas of composition (% vol.): SO_2=16.6, O_2=83.4. Sulfur trioxide is produced at a rate of 8.76 mol/s in 2.72 m^3 of the layer.

Thus, the conditions for the optimal performance of the oxidation process of concentrated sulfide mixtures on such indices as capacity, volume and specific capacity of the layer contact mass have been determined. According to the obtained results in order to perform the process with the maximum productivity in a multi-layer contact plant it is necessary to feed a reactive mixture with the composition (% vol.) SO_2=37.6, O_2=12.8, SO_3=49.6 into every layer. In the special cases when the achievement of the maximum specific capacity or the minimum contact mass volume is desired, it is necessary to feed a reactive mixture free of SO_3 with the composition (% vol.): SO_2=15–17, O_2=83–85 to the layer input. Note that the contact mass volume is 32.7 times less than that required for SO_2 oxidation in a layer performing at the maximum specific capacity, but in this case the produced SO_3 is 30% less than when operating with maximum oxidation capacity.

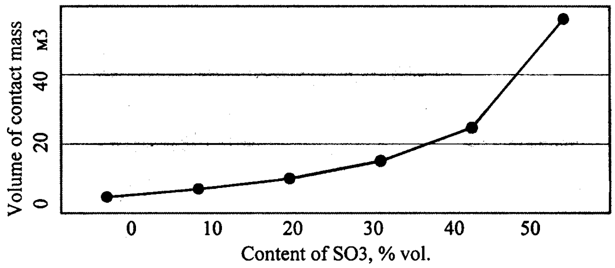
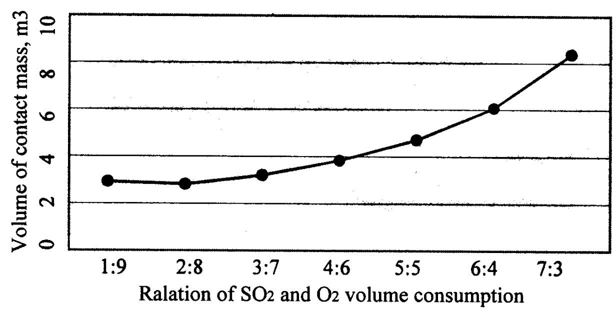


Figure 4. Dependence of the layer volume on the SO_3 content in the reactive gas with 100 mol/s consumption, SO_2:O_2=1:1



Figuere 5. Dependence of the layer volume on the SO_2:O_2 relation in the reactive gas

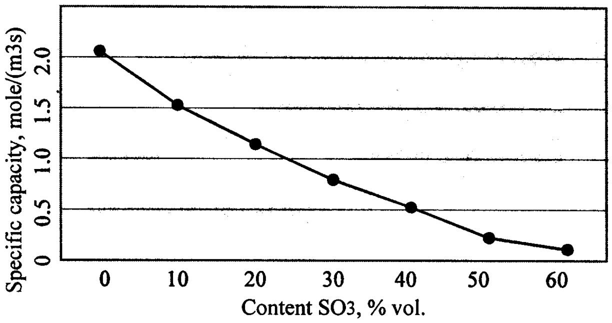


Figure 6. Dependence of the specific capacity on the SO_3 content in the reactive gas of 100 mol/s, SO_2:O_2=1:1

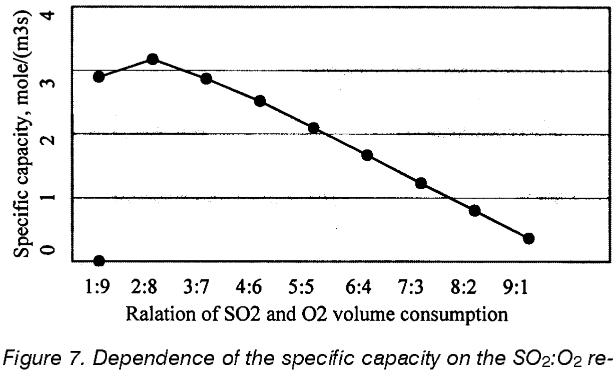


Figure 7. Dependence of the specific capacity on the SO_2:O_2 relation in the reactive gas

Extreme values on such oxidation efficiency indices of the concentrated mixtures such as the contact mass volume and specific capacity of the contact mass volume are given by reactive gases that are similar in composition.

SYMBOLS

C^* – mean integral heat capacity over the interval $[T^0, T^f]$, J/(K · mol);
C – gas heat capacity, J/(K · mol);
K_e – equilibrium constant of the chemical reaction, $Pa^{-0.5}$;
K – reaction rate constant, mole/(s · m^3 · Pa);
L, l – height of the layer of contact mass and current coordinate, m;
G – gas rate consumption, m^3/s;
N, N_i – molar rate of reactive mixture and i^{th} component, mol/s;
$P, P_i, \Delta P$ – total pressure and partial pressure of the i^{th} component, Pa;
Q_r – thermal effect of the chemical reaction, J/mol;
R – gas constant, J/(K · mol);
S – cross–section area of the contact mass layer, m^2;
$T, \Delta T$ – temperature and temperature difference, K;
X – fractional conversion of SO_2 into SO_3;
V – volume of the layer contact mass, m^3;
r – chemical reaction rate, mol/(m^3 · s);
τ – contact time of the reactive mixture with the catalyst, s;
Δ – constant, K;
ν_i – stoichiometric coefficient of the reaction for the i^{th} component;
Ω – capacity of contact mass layer, mol/s;
ω – specific capacity, mol/(m^3 · s).
i – $\in \{ SO_2, O_2, SO_3 \}$

Superscripts

f – at layer output;
0 – at layer input;
th – theoretical value;
tr – true value;
e – equilibrium value;
p – preset value.

Subscripts

i – ignition of oxidation reaction;
d – thermal catalyst deactivation;

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IZVOD

KONTAKTNA OKSIDACIJA KONCENTROVANIH SMEŠA SUMPOR DIOKSIDA – I

(Naučni rad)

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U radu se analizira katalitički proces oksidacije sumpor dioksida u višeslojnom adijabatskom reaktoru koji uključuje i ciklični tok reakcione smeše. Oksidacija SO_2 kiseonikom u kontaktu sa katalizatorom je analizirana na bazi definisanog matematičkog modela. U prvom delu ovog rada ispitivan je uticaj sastava reakcione smeše na efikasnost procesa oksidacije.

Ključne reči: Oksidacija sumpor dioksida • adijabatski proces • višeslojni reaktor • optimizacija •
Key words: Oxidation of sulfur dioxide • Adiabatic process • Multi layer bed • Optimization •