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*Ya.A. Kalymon, A.B. Helesh, A.V. Slyuzar, Z.O. Znak***THEORETICAL STUDIES OF H₂S, SO₂ AND O₂ ABSORPTION IN MASS EXCHANGED APPARATUS WITH A CONTINUOUS BUBBLING LAYER AND MECHANICAL DISPERSION OF AN ABSORBENT****Lviv Polytechnic National University, Lviv, Ukraine**

The paper reports the results of theoretical studies on kinetic and technological characteristics of oxygen, hydrogen sulfide and sulfur(IV) oxide absorption by water in mass-transfer apparatus of two various types: absorber with a continuous bubbling layer (ACBL) and horizontal absorber with bucket-shaped dispersers (HABD). The main purpose of these studies was the theoretical reasoning for the choice of mass-transfer equipment used in the purification of exhaust gases from sulfur compounds. It was shown that the resistance of the gas phase in ACBL is less than the resistance of the liquid phase by $(2.75...3.78) \cdot 10^6$, $(3.43...4.78) \cdot 10^4$, and $(2.18...3.05) \cdot 10^3$ times for oxygen, hydrogen sulfide, and sulfur oxide, respectively; and it can be neglected when calculating the mass transfer coefficient. It was found that an increase in the diameter of the bubble from $1.5 \cdot 10^{-3}$ m to $3.0 \cdot 10^{-3}$ m leads to a decrease in the mass transfer coefficient by 1.63 times. It was calculated that the values of mass transfer coefficients in ACBL for H₂S and SO₂ are much higher than for O₂, so the purification process will be controlled by the rate of oxygen absorption. It was found that the gas phase resistance during the absorption of oxygen and hydrogen sulfide for HABD has virtually no effect on the mass transfer coefficients and can be neglected. For the absorption of gases that are low-concentrated with respect to sulfur(IV) oxide, the mass transfer coefficient in HABD is determined by the resistance of the gas phase, and the optimal droplet diameter is within 2.0 ± 0.5 mm. It was calculated that the technologically expedient initial speed of dispersed drops is 12 m/s, and their diameter is 1.5...3.0 mm for the industrial HABD. It was shown that the values of kinetic and technological indicators of mass transfer in HABD are several orders of magnitude higher than those in ACBL and have a significant potential for intensification.

Keywords: absorption, oxygen, hydrogen sulfide, sulfur(IV) oxide, water, mass transfer coefficient, mass transfer resistance, mass-transfer apparatus.

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Introduction

Sulfur (S) is a fairly common element in the Earth's crust. Most of it is in a chemically bound state in the form of sulfate and sulfide compounds. Oil, natural gas, coal, shale, ore, etc. almost always contain sulfur compounds. As a result of natural chemical and biological processes and in the process of industrial processing of mineral and organic raw materials, sulfur compounds turn into a gaseous state, usually hydrogen sulfide (H₂S) and sulfur(IV) oxide (SO₂).

In modern technologies of complex processing of raw materials, highly concentrated sulfur-containing gases are utilized to obtain valuable products, usually sulfur of various grades, sulfuric acid, etc. The vast majority of industrial exhaust gases are low-concentrated, the content of H₂S and SO₂ in them being 0.1–0.3 vol.%. The use of such gases is technologically difficult and costly. At the same time, these substances are highly toxic, the maximum permissible concentration (MPC) for H₂S (hazard class 2) and SO₂ (hazard class 3) is 0.008 and

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Theoretical studies of H₂S, SO₂ and O₂ absorption in mass exchanged apparatus with a continuous bubbling layer and mechanical dispersion of an absorbent

0.5 mg/m³, respectively. In addition, when released into the atmosphere, these gases under the action of meteorological factors can form stable acidic aerosols and fall to the Earth's surface in the form of acid rain. Acid rain damages the environment, causes erosion and corrosion destruction of various household structures, etc. Therefore, research aimed at developing technologically efficient and low-cost technologies for the purification of sulfur-containing exhaust gases is relevant and important.

Given the above, the main task in such gases purification is the disposal of harmful components, i.e. their conversion into compounds that do not have harmful effects on the environment, or this effect is negligible. It is advisable to convert these pollutants into sulfur or sulfates, using oxygen in the exhaust gas or air as a cheap oxidant [1,2]. However, the oxidation of H₂S or SO₂ by oxygen in the gas phase is energy-intensive, occurring only at high temperatures usually in the presence of expensive catalysts. Therefore, to reduce the process cost, it is advisable to carry out the oxidation process with oxygen in a liquid medium, in particular in water, because water is a cheap and affordable absorbent [1,2].

In industry, aqueous solutions of catalytically active compounds are alkalized and used to increase the absorption capacity. As catalysts for liquid-phase oxidation of exhaust H₂S- and SO₂-containing gases, various compounds are usually used: iron, cobalt, manganese, chromium, arsenic, vanadium, peroxides, quinones, etc. [1–4]. Despite the significant variety of catalytic liquid-phase methods of gas purification, they are all characterized by three main stages: absorption of contaminants by water (solution); catalytic oxidation of sorbed pollutants, and regeneration of the catalyst by absorbed oxygen. The total rate of the liquid-phase purification process will be determined by the slowest stage, usually the absorption of H₂S, SO₂, or O₂. Therefore, the efficiency of the purification process depends on the choice of technologically substantiated mass-transfer equipment. The most rational mass-transfer apparatus will be an absorber in which the rates of all stages of the process are commensurate, otherwise, it is advisable to spatially separate the stages of the process and conduct them in apparatus that corresponds to the physicochemical essence of each stage.

Various types of mass exchangers for purification of such gases are described in the literature. In particular, bubble-type apparatus operating in foam or bubbling modes are usually used for the purification of sulfur-containing gases [1,3]. In the process of gases purification from H₂S

and SO₂, a horizontal absorber with bucket-shaped dispersants (HABD) [1,2,4,5] has proved itself well. Despite the theoretical and practical value of the research, the results of which are given in publications [1–5], they do not pay enough attention to the theoretical rationale for the choice of absorption equipment for purification of low-concentrated gases from H₂S and SO₂. The above necessitates research that will be a reliable basis for the creation of highly efficient technologies for liquid-phase oxidative purification of sulfur-containing gases.

The aim of the research was to determine the kinetic and technological characteristics of the process of absorption of oxygen, hydrogen sulfide, and sulfur (IV) oxide in apparatus with a continuous bubble layer and a horizontal absorber with bucket-shaped dispersants; and theoretically substantiate the choice of mass-transfer equipment for the processes of liquid-phase oxidation of H₂S and SO₂ by absorbed oxygen.

Experimental

To compare the absorption processes indicators in apparatus with a continuous bubbling layer and mechanical dispersion of the absorbent, it is necessary to mathematically describe the complex processes of mass transfer of moving bubbles and droplets under the same conditions. Therefore, to simplify the calculations, the following general assumptions and boundary conditions were adopted: the absorbent in all studied processes is water; H₂S and SO₂ concentrations in the gas phase are low, and therefore reference values for water and air can be used to describe the physical properties of phases; water droplets and air bubbles have a spherical shape and move vertically from the bottom up, during movement they do not deform, do not pulsate, do not undergo secondary divide or coalescence; gas absorption is an isobaric-isothermal process (P=101325 Pa, T=293 K); and heat transfer between phases is absent. For the calculation of HABD, it was assumed that the process of mass transfer occurs only between gas and «primary» droplets formed by the mechanical dispersion of water.

The analysis and processing of the obtained experimental results were performed using standard Microsoft Office 2016 programs. The least-squares method with an approximation of values by appropriate functions was used to treated mathematical dependences.

Method of calculating the mass transfer coefficients

According to the basic equation of mass transfer, the rate of absorption of the i-th gas can be written as

$$\frac{dn_i}{d\tau} = \frac{1}{(1/\beta_i^g) + (m_i/\beta_i^l)} \cdot F_i \cdot \Delta P_i, \quad (1)$$

where n_i is the amount of the i -th gas (mol); τ is the time (s); β_i^g and β_i^l are the mass transfer coefficients of gas and liquid phases, respectively (mol/(N·s)); m_i is the equilibrium distribution coefficient (phase equilibrium constant) determined by the molar fractions of the i -th component of the gas in air and water; $1/\beta_i^g$ and m_i/β_i^l are the mass transfer resistances from the gas and liquid phases, respectively (N·s/mol); ΔP_i is the driving force of the process (Pa); $\Delta P_i = P_i - P_i^*$ (Pa); P_i is the partial pressure of the i -th component in the gas phase (Pa); P_i^* is the partial pressure of the i -th component in the gas phase above the liquid at equilibrium (Pa); and F_i is the interfacial surface (m²).

Replacing $\frac{1}{(1/\beta_i^g) + (m_i/\beta_i^l)} = Km_i$, we get:

$$\frac{dn_i}{d\tau} = Km_i \cdot F \cdot \Delta P_i, \quad (2)$$

where Km_i is the mass transfer coefficient.

The following equations were used to calculate the mass transfer coefficients:

$$\beta_i^{g'} = \frac{Sh^g \cdot D_i^g}{d_p}; \quad \beta_i^{l'} = \frac{Sh^l \cdot D_i^l}{d_p}, \quad (3)$$

where Sh^g and Sh^l are the Sherwood numbers for gas and liquid phases, respectively; D_i^g and D_i^l are the molecular diffusion coefficients of the i -th component in gas and liquid, respectively (m²/s); and d_p is the particle diameter (bubbles or drops) (m).

The molecular diffusion coefficients of O₂, H₂S, and SO₂ in the air at the temperature of 273 K and pressure of 0.1 MPa are 0.178·10⁻⁴, 0.127·10⁻⁴, and 0.122·10⁻⁴ m²/s, respectively. For temperature T, they were calculated by the following formula:

$$D_i^g = D_i^{g(273)} \cdot \left(\frac{T}{273}\right)^{1.75}. \quad (4)$$

The molecular diffusion coefficients (D_i^l) for O₂, H₂S, and SO₂ in water at the temperature of 293 K are 1.8·10⁻⁹, 1.41·10⁻⁹, 1.47·10⁻⁹ m²/s, respectively.

The recalculation of the values and from m/s into mol/(Pa·m²·s) was carried out according to the following equations [6]:

$$\beta_i^g = \frac{\beta_i^{g'}}{RT}; \quad \beta_i^l = \frac{\beta_i^{l'}}{RT}, \quad (5)$$

where R is the universal gas constant (8.31 J/(mol·K)); and T is the temperature (K).

The values of the phase distribution coefficients (m) were adopted from the calculations given elsewhere [6]; they were $m_{O_2} = 4.05 \cdot 10^9 / 101325 = 39970.4$ and $m_{H_2S} = 4.90 \cdot 10^7 / 101325 = 483.6$ in the H₂O–O₂ and H₂O–H₂S systems, respectively.

SO₂ is a polar compound that is quite soluble in water. Therefore, the relationship between the amount of dissolved gas and its partial pressure is not linear in the wide concentration range of SO₂. At the same time, in the region of low concentrations of SO₂, which are characteristic of exhaust gases, its solubility is described by the Henry's law. Therefore, the following empirical formula [4] was used for approximate calculations of the solubility of SO₂ in water at operating temperatures and partial absorption pressures:

$$C_{SO_2}^l = 2.25 \cdot 10^{-3} \cdot p_{SO_2} \cdot 1.0363^{273-T}, \quad (6)$$

where $C_{SO_2}^l$ is the concentration of SO₂ in the solution (kg/m³); p_{SO_2} is the partial pressure SO₂ (Pa).

The values of the phase distribution coefficient were calculated from Eq. (6) for the system H₂O–SO₂ in the following way:

$$m_{SO_2} = \frac{p_{SO_2} \cdot \rho^l \cdot M_{SO_2}}{C_{SO_2}^l \cdot P \cdot M_{H_2O}} = 966.83 \frac{998.2 \cdot 64}{101325 \cdot 18} = 31.76, \quad (7)$$

where M_{SO_2} and M_{H_2O} are the molecular weights of SO₂ and H₂O, respectively (kg/kmol); and P is the pressure (Pa).

Method of calculating Sherwood numbers for apparatus with a continuous bubbling layer (ACBL)

The calculation of Sherwood numbers for the gas phase during absorption in bubbling apparatus was carried out according to the method proposed elsewhere [6]:

$$Sh^g = \frac{\beta_i^g \cdot d_b}{D_i^g} = \frac{1}{6 \cdot Fo^{g'}} \left[1 - \frac{6}{\pi^2} \exp\left(-4\pi^2 \cdot Fo^{g'}\right) \right]_i, \quad (8)$$

where $Fo^{g'}$ is the Fourier number for the gas phase, which was calculated by the equation

$$Fo^{g'} = \frac{D_i^g \cdot \tau}{d_b^2}, \quad (9)$$

where τ is the time of contact of the bubble with the liquid phase (s).

The contact time was taken to be the distance at which the bubble travels a distance equal to its diameter, i.e. $\tau = d_b/w_b$, where w_b is the bubble floating-up rate (m/s). Then we have:

$$Fo^{g'} = \frac{D_i^g \cdot \tau}{d_b^2} = \frac{D_i^g}{d_b \cdot w_b} = \frac{1}{Pe^{g'}} \quad (10)$$

where $Pe^{g'}$ is the Peclet diffusion number for the gas phase $\left(Pe^{g'} = \frac{d_b \cdot w_b}{D_i^g} \right)$.

Sherwood numbers (Sh^l) in the liquid phase for the apparatus with a continuous bubbling layer were calculated by the following equation [6]:

$$Sh^l = \frac{1.13}{\sqrt{Fo^{l'}}} \quad (11)$$

where $Fo^{l'}$ is the Fourier diffusion number for the liquid phase (calculated similarly to Eq. (10) for the gas phase).

Substituting the expression from Eq. (10) instead of $Fo^{l'}$, we obtain:

$$Sh^l = 1.13 \sqrt{Pe^{l'}} \quad (12)$$

where $Pe^{l'}$ is the Peclet diffusion number for the liquid phase, $\left(Pe^{l'} = \frac{d_b \cdot w_b}{D_i^l} \right)$.

The bubble floating-up rate was calculated by the following formula [7]:

$$w_b = \alpha \left(\beta \frac{2\sigma^l}{d_b \cdot \Delta\rho} + g \cdot d_b \frac{\Delta\rho}{2 \cdot \rho^l} \right)^{0.5} \quad (13)$$

where σ^l is the surface tension of water at the boundary with air (N/m); g is an acceleration of gravity (m/s^2); $\Delta\rho = \rho^l - \rho^g$; ρ^l and ρ^g are water and air densities (kg/m^3); and α and β are some empirical constants.

According to the results given in article [7] for bubbles with $d_b \geq 1.5$ mm, the following values are valid: $\alpha = 0.8944$ and $\beta = 1.625$.

Method of calculating Sherwood numbers for horizontal apparatus with bucket-shaped dispersants

For the HABD absorber, there are no criterion equations for calculating the mass transfer coefficients of the gas and liquid phases. Therefore, to calculate these coefficients, we used the criterion equations

for the processes of liquid dispersion in hollow absorbers. In our opinion, the most similar to the absorption in HABD is the equations of Froessling and Masyuk [6]. Accordingly, to calculate the Sherwood numbers in the gas (Sh^g) and liquid phases (Sh^l), the following equations are proposed:

$$Sh^g = 2 + 0.552 \cdot (Re^g)^{0.5} \cdot (Sc^g)^{0.33} \quad (14)$$

$$Sh^l = 0.65 \cdot Pe^{l'} \cdot \left(1 + \frac{\mu^l}{\mu^g} \right)^{-0.5} \quad (15)$$

where Re^g , Sc^g , and $Pe^{l'}$ are the Reynolds

$Re^g = \frac{d_d \cdot \rho^g \cdot w_d}{\mu^g}$, Schmidt $Sc^g = \frac{\mu^g}{D_i^g \cdot \rho^g}$, and Peclet

$Pe^{l'} = \frac{d_d \cdot w_d}{D_i^l}$ numbers, respectively; μ^l and μ^g are the

coefficients of dynamic viscosity of liquid and gas, respectively (Pa·s); w_d is the velocity of the drop relative to the gas (m/s); and d_d is the diameter of the drop (m).

To calculate the speed of the drop and the height of its rise, we used the method described in ref. [8] with changes that take into account that the drop moves from the bottom to the top. Acceleration at time τ_i was calculated by the following equation:

$$a_i = g \cdot \left(1 - \frac{\rho^g}{\rho^l} \right) + \xi_i \cdot \frac{3 \cdot \rho^g \cdot w_{d(i)}^2}{4 \cdot \rho^l \cdot d_d} \quad (16)$$

The medium resistance coefficient ξ_i is a function of the Reynolds number. As the droplet moves, the values of Re_i will change, so we used a generalized equation:

$$\xi_i = \left(\frac{16}{Re_i} + \frac{2.2}{\sqrt{Re_i}} + 0.32 \right) \cdot \left(\frac{1.5 \cdot \mu^l + \mu^g}{\mu^l + \mu^g} \right) \quad (17)$$

According to Euler's numerical method, for a short time $\Delta\tau = \tau_i - \tau_{i-1}$ the acceleration changes insignificantly, i.e. the motion can be considered as uniformly slow. Then the velocity of the drop and the height of its rise at time $\tau_{(i)}$ were calculated by the following formulae:

$$w_{k(i)} = w_{k(i-1)} - a_{(i-1)} \cdot \Delta\tau \quad (18)$$

$$h_{(i)} = h_{(i-1)} + \frac{w_{k(i-1)} + w_{k(i)}}{2} \cdot \Delta\tau \quad (19)$$

The calculation is performed N times before

reaching $w_{\kappa(i)} = 0$, which corresponds to the maximum height of the drop $h_{(i)} = h_{\max}$.

Accordingly, the time required to lift the drop to a height h_{\max} will be

$$\tau_{\max} = N \cdot \Delta\tau. \tag{20}$$

To ensure sufficient accuracy for practical purposes, the calculation of the time interval (step) took $\Delta\tau=0.001-0.002$ s.

According to previous studies [5,9], a technologically appropriate cross-section of the absorber is a semicircle, which is truncated from the exit of the dispersant from the liquid at an angle $\beta=30...35^\circ$ relative to its level (Fig. 1).

The radius of HABD (R_{abs}) and bucket-shaped dispersant (R_{disp}) are 3.0 m and 1.2 m, respectively, and the speed of the ends of the dispersant (initial drop velocity) $w_{\text{disp}} = 10-12$ m/s.

The distance flew by the drop (h_{abs}) in the industrial absorber (from the edge of the bucket of the dispersant to the wall of the absorber) is equal to

$$h_{\text{abs}} = R_{\text{abs}} - R_{\text{disp}} = 3 - 0.6 = 2.4 \text{ m}. \tag{21}$$

The average droplet velocity in the absorber (w_{abs}) was calculated by the formula

$$w_{\text{abs}} = \frac{h_{\text{abs}}}{\tau_{\text{abs}}} = \frac{2.4}{\tau_{\text{abs}}}, \tag{22}$$

where τ_{abs} is the time of the drop in the absorber (time required to raise the drop to a height of 2.4 m) (s).

The aerodynamic effect of gas on the stability of the droplet was evaluated by the value of the Weber

number $\left(We = \frac{\rho^g \cdot w_d^2 \cdot d_d}{2 \cdot \sigma^l} \right)$ [9]. Droplets begin to

break down if the value of the Weber number exceeds the critical value

$$We_{\text{cr.}} = \frac{1}{2} (Re^g)^2 \cdot \left(\frac{\mu^g}{\mu^l} \right)^2 \cdot \frac{\rho^l}{\rho^g} \cdot S, \tag{23}$$

where S is the criterion of drop stability

$$\left(S = \frac{(\mu^l)^2}{\sigma^l \cdot \rho^l \cdot d_d} \right).$$

Simple unauthorized drop division into 2...4 parts occurs under the following conditions:

$$4 \leq We_{\text{cr.}} \leq 20. \tag{24}$$

Methods of comparing technological indicators of efficiency of use of ACBL and HABD

According to Eq. (2), the mass transfer rate at the same driving force of absorption (ΔP) is determined by the product of two units: the mass transfer coefficient (K) and the interfacial surface (F). Therefore, the values of $K \times F$ products for HABD and ACBL absorbers with the same absorption volume and contact time were compared. In addition, an important technological indicator of absorbers, which determines the overall performance of the gas purification system, is the gas capacity ($W_g, \text{m}^3/\text{s}$).

Calculation of technological indicators of HABD

For comparison, we chose an industrial HABD with a radius of 3 m and a length of 3 m. The distance

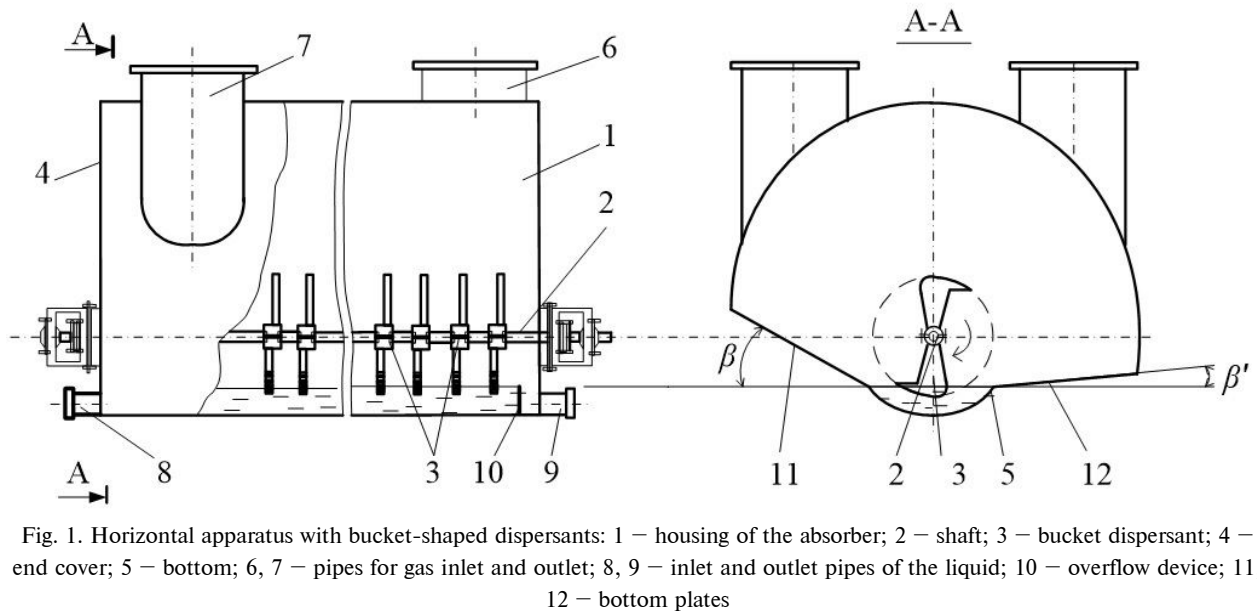


Fig. 1. Horizontal apparatus with bucket-shaped dispersants: 1 – housing of the absorber; 2 – shaft; 3 – bucket dispersant; 4 – end cover; 5 – bottom; 6, 7 – pipes for gas inlet and outlet; 8, 9 – inlet and outlet pipes of the liquid; 10 – overflow device; 11, 12 – bottom plates

between the dispersants was 0.3 m. On the shaft of this absorber, 9 dispersants was placed with a diameter of 1.2 m and a bucket width of 0.09 m. The formation of «secondary» drops was neglected.

The performance of HABD by liquid ($W_{\text{HABD}}^1, \text{m}^3/\text{s}$) was determined by the following formula [9]:

$$W_{\text{HABD}}^1 = \frac{n \cdot 2 \cdot 286.5 \cdot b \cdot v_d \cdot h^{0.9}}{3600}, \quad (25)$$

where n is the number of dispersants (pcs.); $v_d = w_{d(0)}$ is the linear velocity of the ends of the dispersant (m/s); h is the depth of dipping of the dispersant (m), (according to previous studies, $h=0.12$ m).

The interfacial surface F_{HABD} was calculated by the equation:

$$\begin{aligned} F_{\text{HABD}} &= n_d \cdot F_d = \\ &= \frac{W_{\text{HABD}}^1 \cdot (R_{\text{abs}} - R_{\text{disp}}) \cdot 6 \cdot \pi \cdot d_d^2}{w_d \cdot \pi \cdot d_d^3} = \\ &= \frac{W_{\text{HABD}}^1 \cdot (R_{\text{abs}} - R_{\text{disp}}) \cdot 6}{w_d \cdot d_d}, \end{aligned} \quad (26)$$

where n_k is the number of «primary» drops (pcs.) and F_k is the one-drop surface (m^2).

The HABD gas capacity ($W_{\text{HABD}}^1, \text{m}^3/\text{s}$) was calculated in the following way. The velocity of the gas phase in the HABD $v_g=1$ m/s, the cross-section of the apparatus is part of the circle, and the area of which was calculated by the formula $F_{\text{cs}} = 0.5 \cdot \pi \cdot R_{\text{abs}}^2, \text{m}^2$. Then we have:

$$W_{\text{HABD}}^1 = v_g \cdot F_{\text{cs}}. \quad (27)$$

The absorption volume of the industrial HABD will be

$$\begin{aligned} V_{\text{abs}} &= F_{\text{cs}} \cdot t = 0.5 \cdot \pi \cdot R_{\text{abs}}^2 \cdot t = \\ &= 0.5 \cdot 3.14 \cdot 3^2 \cdot 3 = 42.4 \text{ m}^3. \end{aligned}$$

Calculation of technological indicators of ACBL

For comparison, we chose ACBL with an absorption volume $V_{\text{abs}}=42.4 \text{ m}^3$ (cross-sectional area $S=14.13 \text{ m}^2$, and the height of the bubbling layer $h=3$ m). Bubble mode in the apparatus with a continuous bubbling layer takes place at low gas content (up to 0.05). At higher gas contents, coalescence of bubbles occurs and the interfacial surface decreases [6]. Accordingly, the gas-liquid mixture contains $42.4 \text{ m}^3 \cdot 0.05=2.12 \text{ m}^3$ of air.

The interfacial surface of F_{ACBL} was calculated by the following equation:

$$F_{\text{ACBL}} = n_b \cdot F_b = \frac{V_{\text{abs}} \cdot 0.05 \cdot 6 \cdot \pi \cdot d_b^2}{\pi \cdot d_b^3} = \frac{0.3 \cdot V_{\text{abs}}}{d_b}. \quad (28)$$

The capacity of the ACBL for gas ($W_{\text{ACBL}}^g, \text{m}^3/\text{s}$) was calculated by the following equation:

$$W_{\text{ACBL}}^g = \frac{V_{\text{abs}} \cdot 0.05 \cdot w_b}{h}. \quad (29)$$

Results and discussion

Investigation of oxygen, hydrogen sulfide, and sulfur(IV) oxide absorption in the bubbling mode

The results of the calculations of the coefficients and resistances of mass transfer for the gases in the bubbling mode are shown in Table 1. The dependences of the mass transfer coefficients on the

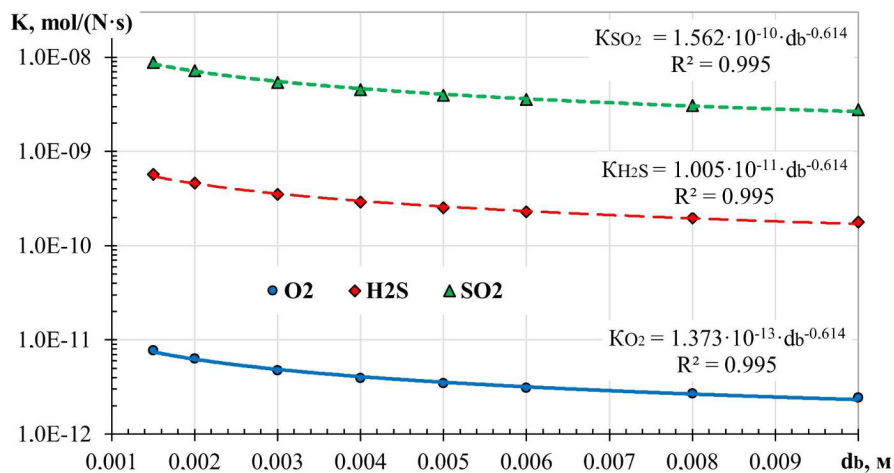


Fig. 2. Dependence of the mass transfer coefficient ($K, \text{mol}/(\text{N}\cdot\text{s})$) on the bubble diameter (d_b, m)

Table 1

The results of calculations of coefficients and resistances of mass transfer in bubbling mode

Gas	Bubbles diameter, $d_b \cdot 10^3$, m	Bubbles rate w_b , m/s	Mass transfer coefficients		Mass transfer resistances		Resistance ratio, l/g
			gas-side, $\beta^g \cdot 10^5$, mol/(N·s)	liquid-side, $\beta^l \cdot 10^7$, mol/(N·s)	gas-side, $1/\beta^g \cdot 10^{-4}$, N·s/mol	liquid-side, m/β^l , N·s/mol	
O ₂	1.50	0.364	2.129	3.098	4.697	$1.290 \cdot 10^{11}$	$2.75 \cdot 10^6$
	2.00	0.321	1.800	2.518	5.557	$1.587 \cdot 10^{11}$	$2.86 \cdot 10^6$
	3.00	0.274	1.434	1.901	6.972	$2.103 \cdot 10^{11}$	$3.02 \cdot 10^6$
	4.00	0.251	1.239	1.576	8.071	$2.536 \cdot 10^{11}$	$3.14 \cdot 10^6$
	5.00	0.240	1.121	1.378	8.917	$2.901 \cdot 10^{11}$	$3.25 \cdot 10^6$
	6.00	0.235	1.046	1.244	9.561	$3.212 \cdot 10^{11}$	$3.36 \cdot 10^6$
	8.00	0.235	0.962	1.077	10.395	$3.711 \cdot 10^{11}$	$3.57 \cdot 10^6$
	10.00	0.241	0.925	0.977	10.814	$4.093 \cdot 10^{11}$	$3.78 \cdot 10^6$
H ₂ S	1.50	0.364	1.945	2.742	5.141	$1.764 \cdot 10^9$	$3.43 \cdot 10^4$
	2.00	0.321	1.635	2.229	6.117	$2.170 \cdot 10^9$	$3.55 \cdot 10^4$
	3.00	0.274	1.296	1.682	7.714	$2.875 \cdot 10^9$	$3.73 \cdot 10^4$
	4.00	0.251	1.119	1.395	8.935	$3.466 \cdot 10^9$	$3.88 \cdot 10^4$
	5.00	0.240	1.015	1.219	9.855	$3.966 \cdot 10^9$	$4.02 \cdot 10^4$
	6.00	0.235	0.949	1.101	10.534	$4.391 \cdot 10^9$	$4.17 \cdot 10^4$
	8.00	0.235	0.880	0.953	11.358	$5.073 \cdot 10^9$	$4.47 \cdot 10^4$
	10.00	0.241	0.854	0.864	11.708	$5.595 \cdot 10^9$	$4.78 \cdot 10^4$
SO ₂	1.50	0.364	1.923	2.800	5.200	$1.134 \cdot 10^8$	$2.18 \cdot 10^3$
	2.00	0.321	1.615	2.276	6.191	$1.396 \cdot 10^8$	$2.25 \cdot 10^3$
	3.00	0.274	1.281	1.717	7.808	$1.849 \cdot 10^8$	$2.37 \cdot 10^3$
	4.00	0.251	1.106	1.425	9.042	$2.230 \cdot 10^8$	$2.47 \cdot 10^3$
	5.00	0.240	1.003	1.245	9.969	$2.551 \cdot 10^8$	$2.56 \cdot 10^3$
	6.00	0.235	0.939	1.124	10.650	$2.825 \cdot 10^8$	$2.65 \cdot 10^3$
	8.00	0.235	0.872	0.973	11.469	$3.263 \cdot 10^8$	$2.84 \cdot 10^3$
	10.00	0.241	0.847	0.883	11.808	$3.598 \cdot 10^8$	$3.05 \cdot 10^3$

diameter of the bubble are shown in Fig. 2.

The calculations based on the criterion equations for the bubble mode in the apparatus with a continuous bubbling layer at low gas content (up to 0.05) have shown the following.

1. The mass transfer resistance of the liquid-side is higher than the resistance of the gas-side by $(2.75...3.78) \cdot 10^6$, $(3.43...4.78) \cdot 10^4$ and $(2.18...3.05) \cdot 10^3$ times during the absorption of oxygen, hydrogen sulfide and sulfur oxide, respectively, (Table 1). The resistance of the gas-side for gases has practically no effect on the mass transfer coefficients and can be neglected. As the bubble diameter increases, the mass transfer support from both the gas and liquid phases increases, which can be explained by a decrease in the bubble velocity with increasing its diameter.

2. The mass transfer coefficients decrease for all gases with increasing bubble diameter (Fig. 2). The obtained dependences are well described by power functions (equations and coefficients of determination (R^2) are given in Fig. 2). Thus, according to the results of calculations, an increase

in the bubble diameter from $1.5 \cdot 10^{-3}$ to $3.0 \cdot 10^{-3}$ m leads to a decrease in the mass transfer coefficient by 1.63 times; while an increase in the bubble diameter from $1.5 \cdot 10^{-3}$ to $10.0 \cdot 10^{-3}$ m leads to a decrease in the mass transfer coefficient by 3.17 times.

3. The values of mass transfer coefficients for oxygen are much lower than for H₂S and SO₂, by 73.2 and 1138 times, respectively. Therefore, the purification process will be limited by the oxygen absorption rate in the apparatus with a continuous bubbling layer. According to Eq. (2), to achieve comparable absorption rates, it is necessary that the oxygen concentration in the gas mixture, which is fed to the purification into ACBL, will be 2 and 3 orders of magnitude higher than the concentration of H₂S and SO₂, respectively.

Investigation of the absorption of oxygen, hydrogen sulfide and sulfur(IV) oxide by a drop of water

In this series of studies, the influence of the values of the initial velocity and diameter of the droplets on the maximum lifting height, contact time, and average droplet velocity was established at the first stage. In addition, the aerodynamic effect of

gas on the stability of the droplet was evaluated using the Weber test. The research was conducted for initial speeds of 10–12 m/s, which according to research [9] are the most technologically feasible for industrial HABD. The results of the calculations are shown in Table 2.

As can be seen from the obtained results (Table 2), the droplets with $d_d \geq 2.5 \cdot 10^{-3}$ m and $d_d \geq 1.5 \cdot 10^{-3}$ m will be formed at the initial velocity of the droplet of 10 m/s and 12 m/s, respectively. An increase in the droplet diameter leads to a significant decrease in the interfacial surface and a decrease in the absorption rate (Eq. (2)). It is impractical to increase the initial velocity of the drop greater than 12 m/s for economic reasons: the specific energy costs for dispersing the liquid will increase [9]. At the speed of 12 m/s and a droplet diameter of $(1.5-3.0) \cdot 10^{-3}$ m, the value of the Weber number does not exceed the critical value,

thus the primary droplets in the process of movement will not be destroyed under the influence of aerodynamic drag. With an increase in the droplets diameter greater than $4 \cdot 10^{-3}$ m, the probability of their division increases. Given the obtained results, further studies were performed for drops with a diameter of $(1.5-3.0) \cdot 10^{-3}$ m at the speed of 12 m/s.

At the second stage, the mass transfer coefficients for droplets with a diameter of $(1.5-3.0) \cdot 10^{-3}$ m at their initial velocity of 12 m/s were estimated. The results of the calculations of the coefficients and resistances of mass transfer in Table 3, and the dependence of the mass transfer coefficients on the diameter of the drops are presented graphically in Fig. 3.

The calculations based on the criterion equations for the mode of mechanical dispersion of the absorbent showed the following.

Table 2

The influence of the initial velocity ($w_{d(0)}$) and the diameter of the droplets (d_d) on the technical indicators of absorption

$w_{d(0)}$, m/s	Indicators	Droplet diameter ($d_d \cdot 10^3$), m						
		1.0	1.5	2.0	2.5	3.0	4.0	5.0
10	h_{max} , m	1.557	2.036	2.396	2.674	2.901	3.247	3.498
	τ_{max} , s	0.463	0.555	0.618	0.666	0.702	0.756	0.794
	τ_{abs} , s	–	–	–	0.432	0.388	0.350	0.332
	w_{abs} , m/s	–	–	–	5.556	6.186	6.857	7.229
	We_{cr}	0.779	1.169	1.558	1.948	2.337	3.116	3.895
12	h_{max} , m	1.832	2.409	2.916	3.302	3.623	4.128	4.509
	τ_{max} , s	0.489	0.574	0.666	0.722	0.768	0.836	0.888
	τ_{abs} , s	–	0.531	0.352	0.310	0.288	0.267	0.255
	w_{abs} , m/s	–	4.520	6.818	7.742	8.333	8.989	9.412
	We_{cr}	1.122	1.683	2.244	2.804	3.365	4.487	5.609

Table 3

The results of calculations of coefficients and resistances of mass transfer in the mode of mechanical dispersion of the absorbent ($w_{d(0)}=12$ m/s)

Gas	Droplet diameter, $d_d \cdot 10^3$, m	Average droplet rate, w_d , m/s	Mass transfer coefficient		Resistances of mass transfer		Resistance ratio, l/g
			gas-side, $\beta^g \cdot 10^5$, mol/(N·s)	liquid-side, $\beta^l \cdot 10^4$, mol/(N·s)	gas-side, $1/\beta^g \cdot 10^{-4}$, N·s/mol	liquid-side, m/β^l , N·s/mol	
O ₂	1.50	4.520	6.881	1.623	1.453	$2.463 \cdot 10^8$	$1.69 \cdot 10^4$
	2.00	6.818	6.979	2.448	1.433	$1.633 \cdot 10^8$	$1.14 \cdot 10^4$
	2.50	7.742	6.528	2.780	1.532	$1.438 \cdot 10^8$	$0.94 \cdot 10^4$
	3.00	8.333	6.108	2.992	1.637	$1.336 \cdot 10^8$	$0.82 \cdot 10^4$
H ₂ S	1.50	4.520	5.397	1.623	1.853	$2.979 \cdot 10^6$	$1.61 \cdot 10^2$
	2.00	6.818	5.498	2.448	1.819	$1.975 \cdot 10^6$	$1.09 \cdot 10^2$
	2.50	7.742	5.152	2.780	1.941	$1.739 \cdot 10^6$	$0.90 \cdot 10^2$
	3.00	8.333	4.826	2.992	2.072	$1.616 \cdot 10^6$	$0.78 \cdot 10^2$
SO ₂	1.50	4.520	5.244	1.623	1.907	$1.957 \cdot 10^5$	10.26
	2.00	6.818	5.345	2.448	1.871	$1.297 \cdot 10^5$	6.93
	2.50	7.742	5.009	2.780	1.996	$1.142 \cdot 10^5$	5.72
	3.00	8.333	4.693	2.992	2.131	$1.061 \cdot 10^5$	4.98

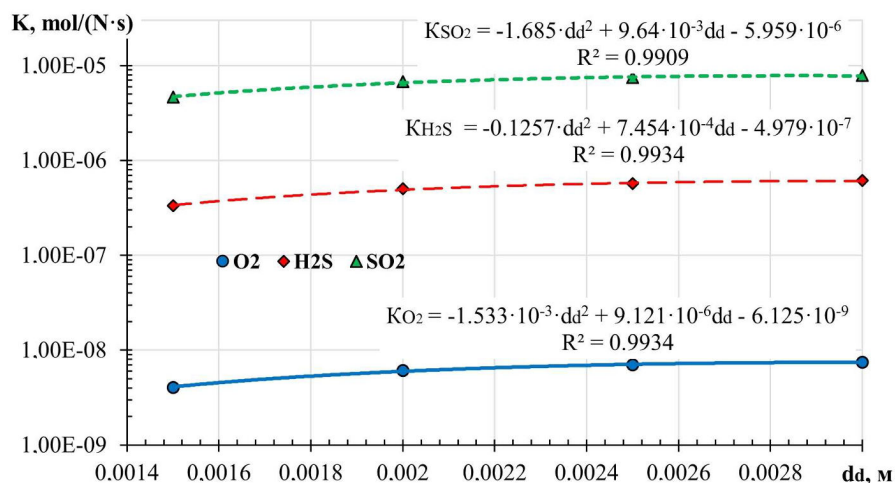


Fig. 3. Dependence of the mass transfer coefficient (K , mol/(N·s)) on the droplet diameter (d_d , m)

1. The resistance of the mass phase of the liquid phase is greater than the resistance of the gas phase by $(0.82...1.69) \cdot 10^4$, $(0.78...1.61) \cdot 10^2$ and $(4.98...10.26)$ times during the absorption of oxygen, hydrogen sulfide and sulfur(IV) oxide, respectively. The resistance of the gas-side during the absorption of oxygen and hydrogen sulfide has virtually no effect on mass transfer coefficients and can be neglected. The resistance of the gas-side during the absorption of sulfur(IV) oxide has a noticeable effect at low concentrations of SO_2 in the gas phase.

2. In the mode of mechanical dispersion of the absorbent (Table 3), in contrast to the bubbling mode (Table 1), the mass transfer resistance decreases with increasing the droplet diameter. This is because as the droplets diameter increases, the average velocity and hence the kinetic energy of the droplet increase (Table 2). The mass transfer resistance of the gas phase generally increases, except for droplets with a diameter of 2 mm, for which the resistance of the gas phase decreases slightly. This non-monotonic nature of the change in resistance is explained by the fact that for drops with a diameter of 2 mm, the increase in the average velocity of the droplet, and accordingly in its kinetic energy is greater than the linear increase in droplet diameter.

3. The mass transfer coefficients increase with increasing the droplet diameter for all gases (Fig. 3). The obtained dependences are well described by polynomial functions (equations and coefficients of determination (R^2) are given in Fig. 3). The most intense relative increase in the mass transfer coefficient (44.7–50.8%) is observed when the droplet diameter increases from 1.5 to 2.0 mm. The relative increase does not exceed 13.5% and 7.6% in the intervals of 2.0 to 2.5 mm and of 2.5 to 3.0 mm,

respectively. Increasing the diameter has a positive effect on the mass transfer coefficient while increasing the diameter of the droplets leads to a decrease in the interfacial surface F (Eq. (2)). This must be taken into account when calculating industrial HABD.

Comparison of technological indicators of efficiency of use of ACBL and HABD in the processes of oxygen, hydrogen sulfide, and sulfur(IV) oxide absorption

To compare the technological indicators of the efficiency of ACBL and HABD, these apparatus with the same absorption volume (42.4 m^3) were chosen. The results of the calculations are shown in Table 4.

As can be seen from the obtained results, HABD is a much more technologically efficient apparatus as compared with ACBL. In particular, at the same driving force of the process, the absorption rate in HABD is 2–3 orders of magnitude higher than in ACBL, and throughput is 54–74 times higher in HABD than in ACBL.

Conclusions

1. The process of liquid-phase oxidative purification of sulfur-containing gases in ACBL is limited by the rate of oxygen absorption. To be able to combine the stage of absorption of these gases and their oxidation, the oxygen concentration in the gas mixture must be 2 and 3 orders of magnitude higher than the concentration of H_2S and SO_2 , respectively. The best technological indicators of absorption in ACBL can be reached at insignificant volume expense of gas and its maximum dispersion.

2. The technologically expedient initial velocity of dispersed droplets is 12 m/s and their diameter is 1.5...3.0 mm for the processes of gases absorption in industrial HABD. For low concentrations of SO_2 gases, the absorption process in HABD is determined

Table 4

Comparison of technological indicators of efficiency of use of ACBL and HABD

Gas	Diameter, $d \cdot 10^3$, m	Mass transfer coefficient		Interfacial surface*		Ratio $K \cdot F_{(HABD)} / K \cdot F_{(ACBL)}$	Gas throughput		Contact time	
		K_{HABD} , mol/(N·s)	K_{ACBL} , mol/(N·s)	F_{HABD} , m ²	F_{ACBL} , m ²		W_{HABD} , m ³ /s	W_{ACBL} , m ³ /s	τ_{HABD} , s	τ_{ACBL} , s
O ₂	1.50	$4.06 \cdot 10^{-9}$	$7.75 \cdot 10^{-12}$	487	8480	30.11	14.13	0.26	0.53	8.24
	2.00	$6.12 \cdot 10^{-9}$	$6.30 \cdot 10^{-12}$	242	6360	37.04	14.13	0.23	0.35	9.36
	2.50	$6.95 \cdot 10^{-9}$	$5.38 \cdot 10^{-12}$	171	5088	43.33	14.13	0.21	0.31	10.24
	3.00	$7.49 \cdot 10^{-9}$	$4.75 \cdot 10^{-12}$	132	4240	49.08	14.13	0.19	0.29	10.95
H ₂ S	1.50	$3.34 \cdot 10^{-7}$	$5.67 \cdot 10^{-10}$	487	8480	33.81	14.13	0.26	0.53	8.24
	2.00	$5.02 \cdot 10^{-7}$	$4.61 \cdot 10^{-10}$	242	6360	41.48	14.13	0.23	0.35	9.36
	2.50	$5.69 \cdot 10^{-7}$	$3.94 \cdot 10^{-10}$	171	5088	48.43	14.13	0.21	0.31	10.24
	3.00	$6.11 \cdot 10^{-7}$	$3.48 \cdot 10^{-10}$	132	4240	54.76	14.13	0.19	0.29	10.95
SO ₂	1.50	$4.66 \cdot 10^{-6}$	$8.81 \cdot 10^{-9}$	487	8480	30.37	14.13	0.26	0.53	8.24
	2.00	$6.74 \cdot 10^{-6}$	$7.16 \cdot 10^{-9}$	242	6360	35.84	14.13	0.23	0.35	9.36
	2.50	$7.45 \cdot 10^{-6}$	$6.12 \cdot 10^{-9}$	171	5088	40.84	14.13	0.21	0.31	10.24
	3.00	$7.85 \cdot 10^{-6}$	$5.41 \cdot 10^{-9}$	132	4240	45.25	14.13	0.19	0.29	10.95

Note: * – the value of the interfacial surface for HABD is given for the contact time in the ACBL.

by the resistance of the gas side and the optimal droplet diameter is within 2.0 ± 0.5 mm.

3. HABD is a highly efficient mass transfer apparatus for the processes of liquid-phase purification of sulfur-containing gases. Its kinetic and technological parameters are several orders of magnitude higher than ACBL. In addition, HABD has a significant potential to intensify mass transfer processes by increasing the number of dispersants and their width. The use of ACBL may be economically feasible given low volumetric gas consumption and low oxidation rates of absorbed H₂S and SO₂.

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ТЕОРЕТИЧНЕ ДОСЛІДЖЕННЯ ПРОЦЕСІВ АБСОРБЦІЇ H_2S , SO_2 І O_2 У МАСООБМІННИХ АПАРАТАХ З СУЦІЛЬНИМ БАРБОТАЖНИМ ШАРОМ І МЕХАНІЧНИМ ДИСПЕРГУВАННЯМ АБСОРБЕНТУ

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У роботі наведені результати теоретичних досліджень кінетичних та технологічних характеристик процесів абсорбції кисню, сірководню і сірки(IV) оксиду водою у масообмінних апаратах з суцільним барботажем шаром (АСБШ) і горизонтальному абсорбері з ковшеподібними диспергаторами (ГАКД). Головною метою зазначених досліджень було теоретичне обґрунтування вибору масообмінної апаратури, яку використовують у технологіях очищення викидних газів від сполук сірки. Показано, що в АСБШ опір газової фази є меншим від опору рідинної фази для кисню у $(2,75...3,78) \cdot 10^6$, сірководню у $(3,43...4,78) \cdot 10^4$, сірки оксиду у $(2,18...3,05) \cdot 10^3$ разів і ним можна знехтувати при розрахунку коефіцієнта масопередачі. Встановлено, що збільшення діаметра бульбашки з $1,5 \cdot 10^{-3}$ до $3,0 \cdot 10^{-3}$ м призводить до зменшення коефіцієнту масопередачі у 1,63 рази. Розраховано, що в АСБШ значення коефіцієнтів масопередачі для H_2S і SO_2 є значно більшими, ніж для O_2 , тому процес очищення буде лімітуватись швидкістю абсорбції кисню. Встановлено, що для апаратів з механічного диспергування рідини опір газової фази під час абсорбції кисню і сірководню практично не впливає на коефіцієнти масопередачі і ним можна знехтувати. Для абсорбції низькоконцентрованих за сірка(IV) оксидом газів в ГАКД коефіцієнт масопередачі визначається опором газової фази, а оптимальний діаметр крапель знаходиться в межах $2,0 \pm 0,5$ мм. Розраховано, що в промислових ГАКД технологічно доцільна початкова швидкість диспергованих крапель становить 12 м/с, а їхній діаметр – $1,5...3,0$ мм. Показано, що значення кінетичних і технологічних показників масопередачі у ГАКД на декілька порядків перевершують аналогічні показники у АСБШ та мають значний потенціал для інтенсифікації.

Ключові слова: абсорбція, кисень, сірководень, сірка(IV) оксид, вода, коефіцієнт масопередачі, опір масовіддачі, масообмінний апарат.

THEORETICAL STUDIES OF H_2S , SO_2 AND O_2 ABSORPTION IN MASS EXCHANGED APPARATUS WITH A CONTINUOUS BUBBLING LAYER AND MECHANICAL DISPERSION OF AN ABSORBENT

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The paper reports the results of theoretical studies on kinetic and technological characteristics of oxygen, hydrogen sulfide and sulfur(IV) oxide absorption by water in mass-transfer apparatus of two various types: absorber with a continuous bubbling layer (ACBL) and horizontal absorber with bucket-shaped dispersers (HABD). The main purpose of these studies was the theoretical reasoning for the choice of mass-transfer equipment used in the purification of exhaust gases from sulfur compounds. It was shown that the resistance of the gas phase in ACBL is less than the resistance of the liquid phase by $(2.75...3.78) \cdot 10^6$, $(3.43...4.78) \cdot 10^4$, and $(2.18...3.05) \cdot 10^3$ times for oxygen, hydrogen sulfide, and sulfur oxide, respectively; and it can be neglected when calculating the mass transfer coefficient. It was found that an increase in the diameter of the bubble from $1.5 \cdot 10^{-3}$ m to $3.0 \cdot 10^{-3}$ m leads to a decrease in the mass transfer coefficient by 1.63 times. It was calculated that the values of mass transfer coefficients in ACBL

for H_2S and SO_2 are much higher than for O_2 , so the purification process will be controlled by the rate of oxygen absorption. It was found that the gas phase resistance during the absorption of oxygen and hydrogen sulfide for HABD has virtually no effect on the mass transfer coefficients and can be neglected. For the absorption of gases that are low-concentrated with respect to sulfur(IV) oxide, the mass transfer coefficient in HABD is determined by the resistance of the gas phase, and the optimal droplet diameter is within 2.0 ± 0.5 mm. It was calculated that the technologically expedient initial speed of dispersed drops is 12 m/s, and their diameter is $1.5...3.0$ mm for the industrial HABD. It was shown that the values of kinetic and technological indicators of mass transfer in HABD are several orders of magnitude higher than those in ACBL and have a significant potential for intensification.

Keywords: absorption; oxygen; hydrogen sulfide; sulfur(IV) oxide; water; mass transfer coefficient; mass transfer resistance; mass-transfer apparatus.

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