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## MATHEMATICAL MODELING OF THE SULFURIC AC- ID DROPS EVAPORATION

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### Abstract

The evaporation process of sulfuric acid solution has been analyzed in the hollow apparatus of column type under direct contact between acid drops and hot gas. On the basis of the mathematical model the main parameters have been calculated: evaporation time, distance passed by a drop and its evaporation temperature. The mathematic dependencies of evaporation time of water and sulfuric acid drops on their initial diameters and air temperature have been developed. The drop diameter of  $\leq 0.5 \cdot 10^{-3}$  m was found to be necessary to carry out the evaporation with the rate sufficient for the industrial columns. The obtained results are proposed to be used to determine the effect of intensifying parameters on the evaporation process of sulfuric acid waste solutions and development of their recycling technology.

**Keywords:** sulfuric acid, evaporation, mathematical model.

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### 1. Introduction

Sulfuric acid (SA) is an important large-tonnage industrial product. A great bulk of SA weak solutions are obtained at many technological processes, in particular at the pigment titani-

um(IV) oxide production. These solutions have to be eliminated or recovered and reused if it is possible [1, 2]. Therefore the search of rational and economically sound recycling method for hydrolysis sulfuric acid is an urgent problem.

In our previous works [3–5] we grounded that the evaporation process of sulfuric acid waste solutions (SAWS) is the most effective under the direct contact of SA dispersed solutions and hot waste gases. One of the widely used apparatuses where this process is easy to be realized is a hollow vertical sprayer unit of column type. Using such apparatus the liquid is dispersed in the upper part of the column and formed drops evaporate during free fall and contact with gas – heat carrier – which is supplied as a counterflow. Thus, the aim of the work was to calculate theoretically the main parameters of SA drops evaporation during their free fall in the gas flow.

The basis for theoretical calculations is the developed by us mathematical model [6] by means of which the weight, temperature, rate and coordinates of drop may be determined in any moment of time.

## 2. Materials and Methods

While developing the mathematical model we assumed the following: from economic and technological point of view the acid is evaporated from 20 to 70 %  $H_2SO_4$ ; the drop has a spherical shape which is not deformed during a movement; water steam partial pressure around the drop corresponds to an unsaturated state and is determined by drop temperature and acid concentration; moisture content and temperature of gas flow are constant; heat transfer between the phases occurs due to convection and the drop moves vertically top-down [5–7].

The intensity of drop heat-and-mass transfer in the gas flow is determined by thermal resistance of liquid and gas phases. In the most cases the resistance of one of the phases is the determinative one. If the drops of 20% SA evaporate in the air at 293 K, the ratio between optimum enthalpies of liquid and gas phases is  $\frac{c_2 \cdot \rho_2}{c_1 \cdot \rho_1} = \frac{3534 \cdot 1139}{1005 \cdot 1.247} = 3212$ , and between heat conductivities –  $\frac{k_2}{k_1} = \frac{0.503}{0.026} = 19.4$

where  $c_1$  and  $c_2$  – specific heats of air and SA solution, J/(kg·K);  $\rho_1$  and  $\rho_2$  – densities of gas and liquid media, kg/m<sup>3</sup>;  $k_1$  and  $k_2$  – heat conductivity factors of air and acid, W/(m·K) [8]. It means that gas flow resistance is a limiting parameter. Thus, we assume in the model that the thermal resistance is practically absent in the liquid phase; the drop heating or cooling rate is determined by the rate of external heat-and-mass transfer; the temperature of all points of the drop is the same.

Using the developed model we may calculate all above-mentioned parameters by different mathematical methods. For this purpose it is necessary to set the initial conditions (coordinates, rate, temperature and size of the drop, acid concentration, time interval  $\Delta\tau$ ) and use Eqs. (1)–(4) to calculate the changes in rate ( $\Delta v_2$ ), moving ( $\Delta r_2$ ), temperature ( $\Delta T_2$ ) and weight ( $\Delta m_2$ ) of the drop during time.

$$\frac{\Delta v_2}{\Delta\tau} = a_2 = g \cdot \left(1 - \frac{\rho_1}{\rho_2}\right) - \xi \cdot \frac{3 \cdot \rho_1 \cdot u^2}{4 \cdot \rho_2 \cdot d_2} \quad (1)$$

$$\Delta r_2 = v_2 \cdot \Delta\tau \quad (2)$$

$$\frac{\Delta m_2}{\Delta\tau} = \frac{\Phi''}{r_3} \quad (3)$$

$$\frac{\Delta T_2}{\Delta\tau} = \frac{\Phi'}{m_2 \cdot c_2} = \frac{\Phi - \Phi''}{m_2 \cdot c_2} \quad (4)$$

where  $\Phi$  – heat flow from the gas medium toward the acid drop, W;  $\Phi'$  and  $\Phi''$  – parts of the flow consumed for drop heating and evaporating, W;  $m_2$  – drop weight, kg;  $r_3$  – specific heat of water

vaporation, J/kg;  $a_2$  and  $g$  – drop and gravitational acceleration,  $m/s^2$ ;  $u$  – drop rate relative to gas (if it is counterflow, then  $u = v_1 + v_2$ );  $v_1$  and  $v_2$  – gas and drop rates,  $m/s$ ;  $\zeta$  – coefficient of medium resistance;  $d_2$  – drop diameter,  $m$ .

To calculate the parameters of Eqs. (1)–(4) we used the following dependencies:

$$\xi = \left( \frac{16}{Re} + \frac{2.2}{\sqrt{Re}} + 0.32 \right) \cdot \left( \frac{1.5 \cdot \mu_2 + \mu_1}{\mu_2 + \mu_1} \right); \quad Re = \frac{d_2 \cdot \rho_1 \cdot u}{\mu_1} \quad (5)$$

$$\Phi'' = r_3 \cdot \pi \cdot d_2^2 \cdot K_D \cdot \frac{M_3}{R} \cdot \left( \frac{p_{30}}{T_1} - \frac{p_{3\infty}}{T_2} \right) \quad (6)$$

$$K_D = \frac{Sh \cdot D}{d_2}; \quad Sh = 2 + 0.6 \cdot Re^{0.5} \cdot Sc^{0.33}; \quad Sc = \frac{\mu_1}{D \cdot \rho_1} \quad (7)$$

$$\Phi = K \cdot \pi \cdot d_2^2 \cdot (T_1 - T_2) \quad (8)$$

$$K = \frac{Nu \cdot k_1}{d_2}; \quad Nu = 2 + 0.6 \cdot Re^{0.5} \cdot Pr^{0.33}; \quad Pr = \frac{\mu_1 \cdot c_1}{k_1} \quad (9)$$

where  $k_1$  – heat conductivity factor,  $W/(m \cdot K)$ ;  $Re$ ,  $Nu$ ,  $Sh$ ,  $Sc$ ,  $Pr$  – Reynolds, Nusselt, Sherwood, Schmidt and Prandtl numbers, respectively;  $T_1$  and  $T_2$  – gas and drop temperatures,  $K$ ;  $\mu_1$  and  $\mu_2$  – coefficients of dynamic viscosity of gas and acid,  $Pa \cdot s$ ;  $K_D$  – coefficient of mass diffusion,  $m/s$ ;  $M_3$  – molecular weight of water,  $M_3 = 0.018$   $kg/mol$ ;  $R$  – universal gas constant,  $R = 8.314$   $J/(mol \cdot K)$ ;  $p_{30}$  and  $p_{3\infty}$  – partial pressures of water steam on the drop surface and in the nucleus of gas flow,  $Pa$ ;  $K$  – heat-transfer coefficient,  $W/(m^2 \cdot K)$ ;  $D$  – diffusion coefficient,  $m^2/s$ ; indices: 1 – gas, 2 – acid, 3 – water [6, 9].

### 3. Results

#### 3.1. Calculation methods

The initial conditions for the calculations are: acid concentration and temperature are 20 % and 293 K, respectively; air is a heat carrier which moves as a counterflow relative to the drop motion with the rate of 1  $m/s$ ; air temperature is 373–773 K; air moisture content is  $10.5 \cdot 10^{-3}$   $kg/kg$ ; drop diameter is  $(0.5–2.0) \cdot 10^{-3}$   $m$ . The finite condition is 70 % concentration of acid drop.

The initial conditions were chosen based on the following reasons: the acid initial concentration corresponds to the approximate concentration of SAWS formed during pigment titanium(IV) oxide production; air temperature and moisture content – to the parameters of industrial waste gases; drops size and gas rate – to the characteristics of industrial sprayers and drop catchers.

The acid final concentration is grounded by the production demands for mineral fertilizers. Moreover in Ref. [6] it is stated that from the technological point of view under direct contact with hot gases the SA concentration should not exceed 70 %.

To calculate drop rate and its motion in any moment of time we used Euler numerical method. It means that acceleration is minor for a short period of time  $\Delta\tau$  ( $\Delta\tau = \tau_{(N)} - \tau_{(N-1)}$ ), i.e. the motion could be considered as uniformly accelerated one. Therefore the calculations algorithm is the following:

- 1) acceleration  $a_{2(N-1)}$  for the moment  $\tau_{(N-1)}$  is calculated according to Eqs. (1) and (5);
- 2) at the moment  $\tau_{(N)}$  the drop rate is:

$$v_{2(N)} = v_{2(N-1)} + a_{2(N-1)} \cdot \Delta\tau \quad (10)$$

and a distance passed by the drop is calculated according to Eq. (2)

$$r_{2(N)} = r_{2(N-1)} + \frac{v_{2(N-1)} + v_{2(N)}}{2} \cdot \Delta\tau \quad (11)$$

3) drop weight at the moment  $\tau_{(N)}$  is calculated according to Eqs. (3), (6) and (7)

$$m_{2(N)} = m_{2(N-1)} - \Delta m_{2(N)} \quad (12)$$

4) drop diameter and acid concentrations are:

$$d_{2(N)} = \sqrt[3]{\frac{6 \cdot m_{2(N)}}{\pi \cdot \rho_{2(N)}}}; \quad C_{2(N)} = C_{2(N-1)} \cdot \frac{m_{2(N-1)}}{m_{2(N)}} \quad (13)$$

5) drop temperature at the moment  $\tau_{(N)}$  is calculated according to Eqs. (4) and (6)–(9)

$$T_{2(N)} = T_{2(N-1)} + \frac{(\Phi_{(N-1)} - \Phi_{(N-1)}'') \cdot \Delta\tau}{m_{2(N-1)} \cdot C_{2(N-1)}} \quad (14)$$

The calculations are conducted N times till SA concentration ( $C_{2(N)}$ ) becomes 70%. Time necessary to achieve the mentioned concentration (time of drop evaporation) is:

$$\tau = N \cdot \Delta\tau \quad (15)$$

To achieve the necessary accuracy the time interval (space) is accepted as  $\Delta\tau = 0.005$  s.

To determine the effect of SA concentration on the efficiency of water evaporation from its solutions the similar calculations for pure solvent (water) were carried out. To compare both results the calculations were conducted till the drop weight decreased by 71.43 %, that corresponds to the amount of water evaporated during SA concentration increase from 20 to 70 %.

The values of temperature and SA concentration vary during all calculation process, hence the phases physical properties vary as well, i.e.:

$$k_1, \mu_1, c_1, \rho_1 = f(T_1); \quad c_2 = f(C_2); \quad \mu_2, c_2, \rho_2, p_3 = f(T_2, C_2)$$

Therefore, reference data of the physical values [8] were exchanged for the developed equations [6]. This allows to use them for calculations.

### 3. 2. Calculation Results

The apparatus efficiency is provided by contact time of drop and hot gas necessary to increase the SA concentration from 20 to 70 % and the distance passed by the drop during this period of time. Temperature and size of drop have the essential effect on the process; the results of corresponding calculations are represented in **Tables 1, 2**.

**Table 1**

Evaporation time of the drop free fallen in the hot air flow ( $v_2^0 = 0$  m/s,  $v_1 = 1$  m/s)

Air temperature, K	Drop initial diameter, m·10 <sup>3</sup>							
	0.5		1		1.5		2	
	H <sub>2</sub> O	H <sub>2</sub> SO <sub>4</sub>	H <sub>2</sub> O	H <sub>2</sub> SO <sub>4</sub>	H <sub>2</sub> O	H <sub>2</sub> SO <sub>4</sub>	H <sub>2</sub> O	H <sub>2</sub> SO <sub>4</sub>
373	7.99	11.83	20.28	29.97	34.53	51.01	49.18	74.16
473	3.23	4.16	8.31	10.68	14.19	18.24	20.66	26.54
573	1.90	2.37	4.97	6.17	8.52	10.56	12.42	15.39
673	1.31	1.60	3.46	4.23	5.95	7.25	8.68	10.58
773	0.98	1.20	2.65	3.20	4.57	5.52	6.67	8.06

**Table 2**

Distance passed by the drop during evaporation time ( $v_2^0 = 0$  m/s,  $v_1 = 1$  m/s)

Air temperature, K	Drop initial diameter, m·10 <sup>3</sup>							
	0.5		1		1.5		2	
	H <sub>2</sub> O	H <sub>2</sub> SO <sub>4</sub>	H <sub>2</sub> O	H <sub>2</sub> SO <sub>4</sub>	H <sub>2</sub> O	H <sub>2</sub> SO <sub>4</sub>	H <sub>2</sub> O	H <sub>2</sub> SO <sub>4</sub>
373	6.20	11.28	47.98	82.57	126.45	210.31	231.50	389.43
473	2.47	4.01	21.43	31.65	56.76	82.48	107.02	154.42
573	1.38	2.18	13.12	18.72	35.67	50.01	68.13	94.79
673	0.88	1.37	9.10	12.80	25.39	35.03	49.15	67.24
773	0.62	0.97	6.98	9.74	20.00	27.39	39.27	53.29

The main parameters affecting the evaporation time (**Table 1**) are the drop initial diameter ( $d_{20}$ ) and air temperature ( $T_1$ ). Time of water evaporation is considerably less than that of SA solution. This difference is essential if air temperatures are low. So, at 773 K the difference between evaporation times is approximately 21 % and at 373 K – 48 %.

For the convenience of practical application the results were mathematically processed and Eqs. (16)–(17) were obtained using the least-squares method with values approximation by power functions.

$$\tau^{H_2O} = \frac{7 \cdot 10^7}{(T_1 - 273)^{1.28}} \cdot d_{20}^{1.33}; \quad (16)$$

$$\tau^{H_2SO_4} = \frac{18 \cdot 10^7}{(T_1 - 273)^{1.4}} \cdot d_{20}^{1.33} \quad (17)$$

The relative error of the above calculations does not exceed 6 %.

The evaporation time of water and SA is proportional to the drop initial diameter to the power of 1.33. The temperature effect is ambiguous: with the increase in air temperature the evaporation time decreases, especially for the SA solutions at high temperatures.

The increase in evaporation time of SA drop is caused by less value of water steam partial pressure over SA solution compared with that over water that results in decreased intensity of water evaporation from SA drop and increased evaporation time.

Frenckel theory of translation motion [10] represents another explanation of temperature effect on water evaporation process. According to this theory the molecules of liquid inside the complexes (cells) formed by neighboring molecules carry out hundreds of oscillatory movements during their "settled life" time until free energy of the complex increases by the magnitude of activation energy under which the molecule performs the active jump. Time of the "settled life" ( $\tau'$ ) is determined by Eq. (18):

$$\tau' = \tau_0 \cdot e^{\frac{E_A}{R \cdot T_2}} \quad (18)$$

where  $\tau_0$  – oscillation period of the molecule that characterizes average duration of the molecule in the "settled" state, s;  $E_A$  – activation energy, J/mol.

With the increase in temperature the "settled life" time of the molecule decreases and the probability that the molecule leaves the liquid increases. It means that an evaporation rate is proportional to the probable escape of the liquid molecules into the gaseous phase. Thus,

$$\frac{\Delta m_2}{\Delta \tau} \sim \frac{1}{e^{\frac{E_A}{R \cdot T_2}}} = e^{-\frac{E_A}{R \cdot T_2}} \quad (19)$$

Taking into account that the activation energy of the sulphuric acid is higher than that of the pure water, it follows from Eq. (19) that it is necessary to increase the temperature of H<sub>2</sub>SO<sub>4</sub> to

overcome the activation barrier. In its turn, the temperature of the solution increases with the increase in air temperature, therefore the effect of air temperature for the SA solutions is higher compared with the effect for water.

It is economically advisable that SA evaporates during single pass through the vertical column, therefore the distance passed by the drop during evaporation will determine the column height. However from **Table 2** it is obvious that within 373–773 K the acid evaporation is possible only for the drops with the diameter of  $\leq 0.5 \cdot 10^{-3}$  m. In other cases economic and design problems occur during manufacturing bulky and material-consumption column, because its height should be within 10–400 m.

Thus, evaporation process in the industrial column apparatus may be realized only by cyclic scheme, with multiple feed of SA for evaporation, which requires additional energy consumption and complicates the process due to SA corrosion activity and necessity of circulating vessel.

The important parameter for technological process and equipment design is the temperature achieved by SA during evaporation. The results are represented in **Table 3**.

**Table 3**

Temperature achieved by the drop during evaporation ( $v_2^0 = 0$  m/s,  $v_1 = 1$  m/s)

Air temperature, K	H <sub>2</sub> O	H <sub>2</sub> SO <sub>4</sub>
373	304.9	349.3
473	315.6	367.3
573	321.9	376.3
673	326.1	381.9
773	328.8	385.7

The values of SA drops final temperature are considerably higher compared with water but in all cases they are lower than the temperatures of water and 70 % H<sub>2</sub>SO<sub>4</sub> boiling (373 and 438 K, respectively) [8]. Such regularity may be explained using Eq. (19). Work function of the water molecules into gas medium from H<sub>2</sub>SO<sub>4</sub> is higher than that of pure water and increases with the increased SA concentration. The difference between activation energies for SA and water should be compensated by increased temperature of acid drop. Hence, overheating is a necessary condition of water evaporation from acid drop. The intensity of evaporation increases with the increase in air temperature.

The temperature of drop evaporated under direct contact with gas-carrier is determined by the temperature of wet thermometer which depends on air temperature, air moisture content and acid concentration. The temperature is not a function of drop diameter and rate. It means that if air parameters are constant ( $T_1, p_{3\infty} = \text{const}$ ) then the drop temperature depends only on acid concentration ( $T_2 = f(C_2)$ ).

**Table 3** shows that at 773 K sulphuric acid achieves maximum temperature of 385.7 K which is lower than the temperature of acid thermal decomposition (403 K). Thus, only water steam transfers to the gas phase from the solution. The absence of SA thermal decomposition renders impossible the formation of sulphoacid fog which is heavy to be trapped and increases the corrosion activity of technological gases.

#### 4. Conclusions

1. Using the developed mathematical model we calculated the time necessary for the contact between acid drop and hot gas. It was determined that evaporation time is proportional to the drop initial diameter to the power of 1.33. Overheating is a necessary condition for water evaporation from acid drops, therefore the final temperature of SA drops is considerably higher compared with water but lower than the temperature of SA thermal decomposition (403 K).

2. The high intensity of SA evaporation in the sprayers of column type may be achieved only for the drops with the diameter of  $\leq 0.5 \cdot 10^{-3}$  m resulting in significant carryover of drops and increased demands for drop catchers and corrosion resistance of the equipment. Therefore the possible way to increase the process intensity is to change the technological regime of evaporator and design of the main apparatus.

3. To determine the influence of the intensifying parameters which allow to select the evaporator regime, to design the main apparatus and to minimize the corrosion effect of SA it is necessary to carry out the additional theoretical and practical investigations, the results of which will be considered in the further publications.

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## DEVELOPING PROCEDURE DETERMINING ICE FORMATION FOR EVALUATION FROST CONCRETE

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#### Abstract

The paper discusses the basic stages of modeling the processes of ice formation concrete. The conditions for the formation of ice in the pore space of concrete. The technique of determining ice formation, based on the finite element method, differential scanning calorimetry, heat and mass transfer processes, the nature of the pore space, which minimizes time determining frost resistance of concrete for general purpose and assess their durability.

**Keywords:** concrete, differential scanning calorimeter, frost-resistance, ice formation, methods

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#### 1. Introduction

В современных условиях оценить морозостойкость бетонов без длительных и дорогостоящих экспериментов возможно по показателю льдообразования [1].

В целом морозостойкость бетона можно рассматривать как накопление напряжений, вызванных циклами объемных деформаций бетона в результате замерзания в его порах воды при температурах ниже 0°C, до начала разрушения бетона. В реальных условиях строи-