Results of Mathematical Modeling of Evaporation Process the Drops of Sulfuric Acid in the Gas Flow

Uliana Khimka[1], Andriy Helesh[2], Roksoliana Bukliv[2], Mariusz Śniadkowski[3*]

[1] Department of Mathematics, Lublin University of Technology, ul. Nadbystrzycka 38A, 20-618 Lublin, Poland
[2] Department of Chemistry and Technology of Inorganic Substances, Lviv Polytechnic National University, Bandera Str. 12, 79013 Lviv, Ukraine
[3] Fundamentals of Technology Faculty, Lublin University of Technology, ul. Nadbystrzycka 36, 20-618 Lublin, Poland
* Corresponding author e-mail: m.sniadkowski@pollub.pl

ABSTRACT

The processes of concentration of dilute solutions of sulfuric acid (H_2SO_4), in particular hydrolytic sulfuric acid for the production of pigment titanium (IV) oxide, were analyzed. It is proposed to concentrate solutions of H_2SO_4 by evaporation in direct contact of acid droplets with hot industrial exhaust gases. The mathematical model of the evaporation of H_2SO_4 drops in a hot gas stream, which used for the calculations, makes it possible to calculate the mass, temperature, velocity, and coordinates of the drop at any time with sufficient accuracy. However, the calculations are difficult, cumbersome and require multiple processing of large data sets. Therefore, the aim of the article was to approximate the calculated technological parameters of sulfuric acid droplet evaporation by obtaining simple mathematical dependencies. The mathematical dependences of the mass transfer coefficient and the distance traveled by a drop of H_2SO_4 during evaporation on air temperature and drop diameter were obtained. It has been established that technologically expedient evaporation of H_2SO_4 drops with a diameter of $\leq 0.5 \cdot 10^{-3}$ m in traditional devices leads to significant droplet loss, increased corrosion of equipment, etc. So, in order to practically implement the technology of evaporation of HSA solutions with industrial exhaust gases, it is necessary to change the technological mode of operation of the evaporator and the design of the main device. It is proposed to use the obtained results to study the methods of intensification of the evaporation process, selecting a modern mass transfer apparatus, developing a technology for utilizing hydrolytic sulfuric acid and producing pigment titanium (IV) oxide.

Keywords: evaporation, hydrolytic sulfuric acid, exhaust gases, mathematical model.

INTRODUCTION

Many technological processes in the chemical and related industries produce significant amounts of dilute sulfuric acid solutions (H_2SO_4). In particular, in the sulfate method of producing pigment titanium (IV) oxide (TiO_2), sulfuric acid, being a necessary raw material, does not pass into the final product, but it is diluted and contaminated during the multi-stage transformation process. In total, 5-6 tons of hydrolytic sulfuric acid (HSA) are formed in the process of producing 1 ton of TiO_2.

The annual global production of pigment TiO_2 reaches 8.4 million tons, including 120 thousand tons of TiO_2 produced in the Ukraine [1]. More than half of the world's TiO_2 production capacity is based on the sulphate method, and in Ukraine all TiO_2 is produced using this method. Accordingly, huge quantities of dilute H_2SO_4 solutions are formed. Despite attempts to improve the sulfuric acid production of pigment TiO_2 [2, 3], the problem of HSA utilization has not been completely solved yet.

The low content of H_2SO_4 in HSA (20–26% by weight) and the presence of soluble salts of Ferrum and other metals are an obstacle to its use. As a result, HSA is a large-tonnage production waste that many factories do not utilize. This

leads to an irreversible loss of valuable chemical raw materials - H_2SO_4. Furthermore, titanium (IV) oxide pigment producers pay significant environmental fines for discharging hydrolytic sulfuric acid. Thus, the production of pigmented TiO_2 carries significant economic losses, which leads to a deterioration in environmental and technical and economic performance. So, the search for a rational, economical method of utilization of hydrolytic sulfuric acid is an extremely urgent issue.

Concentration of HSA to the standard of a marketable product (92.5–94% H_2SO_4, no more than 0.02% Ferrum compounds) is technologically complex and costly. Today, the main method of utilizing the diluted sulfuric acid solutions is to concentrate them by evaporation. The possibility of concentrating aqueous solutions of sulphuric acid by evaporation is based on the fact that up to a concentration of 98.3% H_2SO_4, the ratio of H_2SO_4: H_2O in the gas phase is lower than in the liquid. Therefore, theoretically, the concentration of sulphuric acid can be increased to 98.3% H_2SO_4 by simple evaporation.

At the beginning of evaporation, only water vapour passes into the gas phase, so the acid concentration increases proportionally. This process continues until the H_2SO_4 content in the solution is close to 80%. When heated further, sulfuric acid also passes into the gaseous state along with water vapour. Thus, at atmospheric pressure, the boiling point of 85% sulfuric acid is 227 °C, and the content of H_2SO_4 in saturated steam is 0.14%; for 95% sulfuric acid, it is 301 °C and 40.8%; for 98% sulfuric acid, it is 332 °C and 85.0%, respectively. In addition, during the evaporation of sulfuric acid, its thermal decomposition is possible with the subsequent formation of a fine mist, which is difficult to capture. Noticeable thermal decomposition of H_2SO_4 was observed only at temperatures above 130 °C. The degree of thermal decomposition at 200 °C is 2.5; at 300 °C - 27.1; at 400 °C - 69.1% [4]. So, it is advisable to evaporate sulfuric acid solutions at a temperature not exceeding 130 °C.

Traditionally, natural gas has been used as an energy carrier for the evaporation of dilute sulfuric acid solutions. Due to the high price of natural gas, this method is not economically unprofitable. An alternative to natural gas can be the exhaust hot gases, unsaturated with water vapor, which are formed in the production of pigment TiO_2. Works [5-7] theoretically substantiated and practically confirmed the feasibility of evaporation of HSA with hot exhaust gases under direct contact of waste gases - dispersed sulfuric acid solution. A method for implementing the technology of evaporation of hydrolytic sulfuric acid solutions is proposed [8]. Despite the practical value of the results obtained, for the successful implementation of the proposed technology and the possibility of its scaling, it was necessary to create its mathematical model.

As noted above, evaporation is most efficient when dispersed sulfuric acid solutions and hot gases are in direct contact. The HSA dispersion is a totality of droplets of different sizes which move with different velocities in the hot gas flow. It is difficult to describe such a complex dynamic system mathematically. So, in order to simplify, a single drop that evaporates while moving in a gas stream is taken as the object of research.

The evaporation of liquid droplets is a classical problem of the theory of dispersed systems; a significant number of empirical and theoretical investigations have been summarized and systematized in the works of Fuchs et al. [9, 10]. The problem of a formalized description of the process of droplets evaporation has not lost its relevance today. In particular, the authors of [11] modeled the process of evaporation of sulfuric acid particles in the atmospheres of the Earth and Venus, and in works [12, 13] theoretically and experimentally determined the heat and mass transfer coefficients during the evaporation of liquid droplets etc. Thus, despite the large number of publications, there is no rigorous mathematical solution to this problem today; the problem is generally solved by numerical or experimental methods.

In works [14-16], attempts at an approximate solution are relevant only for certain limiting or partial cases of evaporation. In all of these works, the evaporation of droplets of individual liquids (mainly water) was considered, which retain their qualitative and quantitative composition during evaporation, and the properties of the gas and liquid phases were a function of the nature of the components and temperature. In the case of evaporation of sulfuric acid droplets, its concentration will change over time, that is, the properties of the liquid phase will also depend on the time-varying quantitative composition of the droplet.

In work [17], the mathematical model "sulfuric acid droplet - heated gas" is described. The process of heat and mass transfer is described as

a system of differential equations of conservation of momentum, energy, and mass. Experimental data are used for the calculation, usually generalized on the basis of the similarity theory. The proposed model makes it possible to calculate the mass, temperature, velocity, and coordinates of a drop at any given time with sufficient accuracy. At the same time, numerical calculations are cumbersome and require multiple calculations of large data sets, which greatly complicates their practical use to establish a technologically feasible mode of evaporation of the HSA and select an efficient main device. Thus, the aim of this article is to approximate the technological parameters of sulfuric acid droplets evaporation calculated according to the mathematical model. To achieve this goal, it is necessary to solve the following tasks:

- to determine the dependence of the average mass transfer coefficient on the initial diameter and temperature of the heat carrier gas.
- to theoretically investigate the influence of system parameters on the distance traveled by H_2SO_4 droplets and to establish analytical dependencies between them.

MATERIALS AND METHODS

Theoretical model is based on real reference experimental data. The heat carrier gas is air [19][22], the temperature of which varies within 100 °C to 500 °C, and its water content is equal to $10.5 \cdot 10^{-3}$ kg/kg which is average annual value for the city of Armyansk in Crimea. For theoretical calculations, reference values of parameters were used for water solutions of sulphuric acid H_2SO_4 [18], the concentration of acid during the evaporation process increases from 20% to 70%. The developed model "sulfuric acid droplet - heated gas" was used to calculate the evaporation rate [13]. To simplify the calculations, it was assumed that the droplet has a spherical shape and does not deform during movement; the temperature in the entire volume of the droplet is the same, and therefore the rate of its evaporation is determined by the intensity of external heat and mass transfer. Heat is transferred by convection; thermal resistance in the liquid phase is practically absent. The partial pressure of water vapor around the droplet is determined by the temperature of the drop and the content of H_2SO_4 in it; the moisture content and temperature of the gas flow do not change.

The calculations were performed under the following conditions: initial and final acid concentrations - 20% and 70% respectively; the initial temperature of the acid droplets is 20 °C or equal to the calculated temperature of the wet thermometer; the diameter of the droplets varies within 0.5–$2.0 \cdot 10^{-3}$ m; the heat carrier gas is air, its temperature, moisture content and velocity are 100–500 °C, $10.5 \cdot 10^{-3}$ kg/kg and 1 m/s, respectively; the phase movement is contra-flow.

Changes in the velocity (Δv_2), displacement (Δr_2), temperature (ΔT_2) and mass (Δm_2) of a sulfuric acid droplet over time were calculated using the following equations:

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

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

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

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

where: 1 - gas, 2 - acid; 3 - water;
Φ – heat flow from the gas environment to the acid droplet, W;
Φ' i Φ'' – parts of the flow that are spent on heating and evaporation of the droplet, W;
m_2 – mass of the droplet, kg;
c_2 – specific heat of the sulfuric acid solution, J/(kg·K);
r_3 – specific heat of vaporization of water, J/kg;
a_2 and g – accelerations of the drop and free fall, m/s^2;
u – drop velocity relative to the gas (in the case of countercurrent movement, $u = v_1 + v_2$),
v_1, v_2 - gas and drop velocities, m/s;
ξ - coefficient of resistance of the environment; d_2 - drop diameter, m.

The following dependencies were used to calculate the values included in equations (1-4):

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

$$Re = \frac{d_2 \cdot \rho_1 \cdot u}{\mu_1}$$

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

$$K_D = \frac{Sh \cdot D}{d_2};$$

$$Sh = 2 + 0.6 \cdot Re^{0.5} \cdot Sc^{0.33}; \qquad (7)$$

$$Sc = \frac{\mu_1}{D \cdot \rho_1}$$

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

$$K = \frac{Nu \cdot k_1}{d_2};$$

$$Nu = 2 + 0.6 \cdot Re^{0.5} \cdot Pr^{0.33}; \qquad (9)$$

$$Pr = \frac{\mu_1 \cdot c_1}{k_1}$$

where: *Re, Nu, Sh, Sc, Pr* – Reynolds, Nusselt, Sherwood, Schmidt, Prandtl criteria, respectively;

μ_1 and μ_2 – coefficients of dynamic viscosity of gas and acid, Pa·s;

T_1, T_2 – temperatures of gas and droplet, K;

K_D – mass diffusion coefficient (mass transfer coefficient), m/s;

M_3 – molecular weight of water, $M_3 = 0.018$ kg/mol;

R – universal gas constant, $R = 8.314$ J/(mol·K);

p_{30} and $p_{3\infty}$ – partial pressures of water vapor on the surface of the droplet and in the core of the gas flow, Pa;

K – heat transfer coefficient, W/(m²·K);

k_1 – thermal conductivity of gas, W/(m·K);

D – diffusion coefficient, m²/s.

Euler's numerical method was used to calculate the velocity of the droplet and its displacement at any given time:

1) using formula 1, calculate the acceleration $a_{2(N-1)}$ at time $\tau_{(N-1)}$;

2) at time $\tau_{(N)}$ the velocity of drop will be

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

3) the distance travelled by the droplet (formula 2) will be

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

4) the mass of the droplet at time $\tau_{(N)}$ will be

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

5) using formula 4, calculate the temperature of droplet at time $\tau_{(N)}$

$$T_{2(N)} = T_{2(N-1)} + \frac{(\Phi_{(N-1)} - \Phi''_{(N-1)}) \cdot \Delta\tau}{m_{2(N-1)} \cdot c_{2(N-1)}} \qquad (13)$$

Using formulas below, calculate drop diameter and concentration of acid

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

$$C_{2(N)} = C_{2(N-1)} \cdot \frac{m_{2(N-1)}}{m_{2(N)}} \qquad (14)$$

The calculation is repeated *N* times until the concentration of sulfuric acid $C_{2(N)} = 70\%$. The time interval (step) is $\Delta\tau = 0.005$ s.

The productivity of the evaporator will be determined by the evaporation rate of the droplets in it. In general, the evaporation rate of droplets is described by the formula

$$I = K_D \cdot \Delta C_3 \cdot f_2 \qquad (15)$$

where: ΔC_3 – difference in water vapor concentration (the driving force of the process), kg/m³;

f_2 – heat transfer surface (droplet surface, $f_2 = \pi \cdot d_2^2$), m².

From the foregoing, it can be seen that the main factors that determine the evaporation time are: gas temperature; droplet diameter; heat and mass transfer coefficients, which are functions of system parameters and relative droplet velocity. Thus, if the gas parameters are stable, the factors affecting the "sulfuric acid droplet - heated gas" system are the temperature, diameter, and velocity of the droplet.

RESULTS AND DISCUSSION

To establish the influence of hydrodynamic conditions on the evaporation process, the average values of the droplet velocity relative to the gas were calculated [20]. Drops fall in a gas environment with an initial velocity of $v_2 = 0$ m/s, and the gas moves against them with a counterflow of $v_1 = 1$ m/s. That is, the initial relative velocity of the drop is $u_m = v_2 + v_1 = 1$ m/s. The results of the calculations are shown graphically in Figure 1.

As can be seen from the graph, the average velocity of the droplets increases significantly with the increase in the droplet diameter. This is due to the fact that as the droplet diameter increases, the resulting gravity and resistance of the medium increases, since the mass of the droplet increases in a cubic and the cross-sectional area in a quadratic dependence.
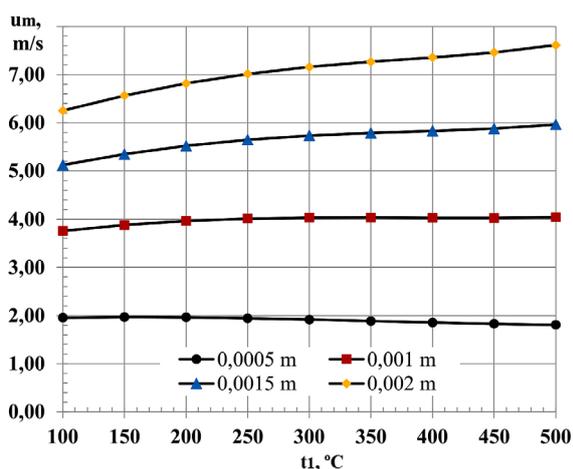
Fig. 1. Dependence of the relative average droplet velocity (u_c, m/s) on their initial diameter (d_{20}, m) and gas temperature (t_1, °C)

Fig. 2. Dependence of the average mass transfer coefficient (K_D, m/s) on the initial diameter of the acid droplet (d_{20}, м) and gas temperature (t_1, °C)

As the temperature increases, the average velocity of large droplets increases as a result of a decrease in the density of the gas environment and, consequently, a decrease in its resisting force. For small drops ($d_{20} = 0.5 \cdot 10^{-3}$ m), an increase in temperature leads to a decrease in velocity, which may indicate a high intensity of evaporation of small drops, and a corresponding decrease in the kinetic energy of the drop.

In general, as the diameter of a droplet increases, its relative velocity increases, which intensifies mass transfer processes. On the other side, large droplets have a smaller specific evaporation surface, which reduces their evaporation rate. An indicator that takes these two factors into account is the mass transfer coefficient (K_D, m/s). Therefore, the next stage of the investigation was to determine the effect of the initial droplet diameter and gas temperature on the average K_D value. The results are shown graphically in Figure 2.

With increasing temperature, the KD value increases directly, especially for small droplets ($d_{20} = 0.5 – 0.1 \cdot 10$ m). This is due to the temperature increase in the diffusion coefficient of water vapor in air D.

For the convenience of practical use, the results obtained were mathematically processed by the least squares method with approximation of values by step functions

$$K_D = \frac{5.34 \cdot 10^{-5}}{d_{20}^{0.41}} \cdot \frac{3.64 \cdot 10^{-2}}{d_{20}^{0.29}} \qquad (16)$$

The relative calculation error within the investigated limits does not exceed 1.8%.
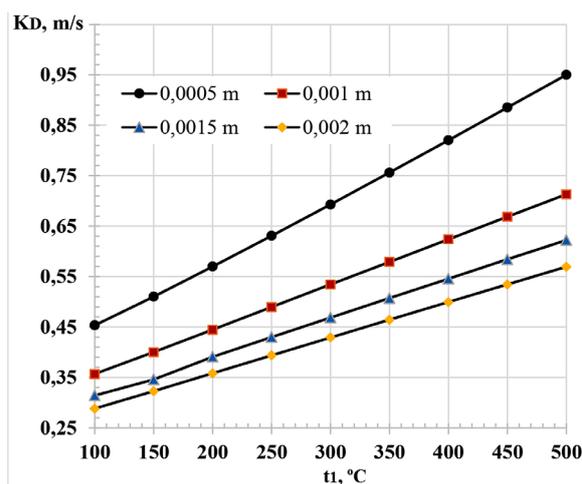
For the practical implementation of the sulfuric acid evaporation technology, it is important to select heat and mass transfer equipment that matches the physicochemical nature of the process [21]. It is economically viable that the evaporation of sulfuric acid from the initial (20%) to the final (70%) concentration occurs in one passage of droplets through the apparatus. Therefore, the distance traveled by a drop during evaporation will determine the type and dimensions of the evaporator. The results of calculations of the distance traveled by a droplet at different initial diameters and gas temperatures are shown in Figure 3.

As can be seen from the figure, in the temperature range of 100-500 °C, it is practically possible to realize the evaporation of acid in one pass only for droplets with a diameter of $\leq 0.5 \cdot 10^{-3}$ m, and in other cases there are constructive and economic difficulties in the manufacture of large and material-intensive evaporating equipment, since, according to the calculations (Fig. 3), the distance traveled by the droplet will be in the range of ~10–390 m. So, for droplets with a diameter of $1.0\text{-}2 \cdot 10^{-3}$ m, the evaporation process can be realized according to a cyclic scheme, with multiple supply of acid solution for evaporation [23]. Such a scheme of evaporation requires additional energy costs for dispersing and supplying sulfuric acid solutions, as well as the use of additional circulation tanks.

For ease of practical use, the results of the calculations were mathematically processed and, using the least squares method with approximation of values, the following dependencies were derived:
- for droplets with a diameter of $d_{20} = 0.5 \cdot 10^{-3}$ m, the distance is calculated by the formula
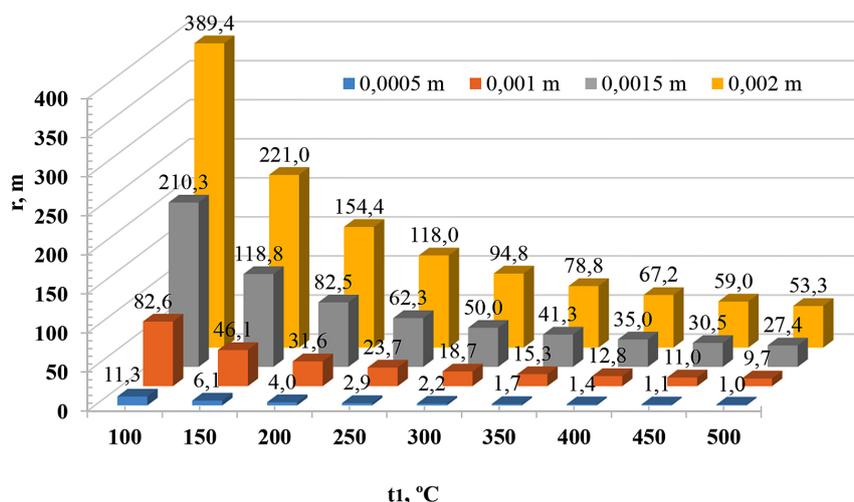
Fig. 3. Dependence of the distance traveled by a droplet (г, m) on its initial diameter (d_{20}, м) and gas temperature (t_1, ºC)

$$r = \frac{1.25 \cdot 10^4}{t_1^{1.52}} \qquad (17)$$

- for droplets with a diameter of $1.0 \dots 2.0 \cdot 10^{-3}$ m - by the formula

$$r = \frac{2.68 \cdot 10^{10}}{t_1^{1.22}} \cdot d_{20}^2 - \frac{1.15 \cdot 10^3}{t_1^{0.90}} \qquad (18)$$

The relative calculation error within the specified concentration and temperature limits according to Formula 17 does not exceed 2%, and according to Formula 18 - 5%.

CONCLUSIONS

The obtained analytical dependences made it possible to scientifically substantiate the choice of heat and mass exchange equipment for the technology of concentration of waste solutions of HSA production of pigment TiO_2. Practically, evaporation of HSA in traditional column devices is possible only for droplets with a diameter of $\leq 0.5 \cdot 10^{-3}$ m. The implementation of such a process will lead to significant droplet loss, increased corrosion of equipment, etc. Therefore, possible methods of implementing the technology of evaporation of HSA solutions with exhaust industrial gases require a change in the technological mode of operation of the evaporator and the design of the main apparatus. To establish the technologically feasible operation mode of the evaporator and select an efficient main device, it is necessary to conduct additional theoretical and experimental studies.

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