

The article establishes quantitative correlations for processes of heterogeneous chemical interaction between a solid body and a liquid reagent with liberation of gas under conditions of free convection.

Chemical boiling is the name for processes of heterogeneous chemical interaction between a solid body and a liquid reagent, as a result of which one of the products is released in the form of gas [1]. Such processes occur when carbonates, sulfides, phosphites are decomposed by acids or when metals interact with acids. Bubbles form directly on the solid surface, grow and detach themselves, and they act on the diffusion boundary layer of the liquid.

Mass transfer under conditions of free convection was studied on the example of the dissolution of spherical particles of magnesium and calcite in different acids. The method of carrying out such experiments was described by Zelin'ski [2].

Between mass transfer in chemical boiling and heat transfer in boiling there is an analogy whose principal aspects were dealt with by Aksel'rud and Molchanov [3]. We point out an analogous kind of dependence of the mass-transfer coefficient  $k$  on the motive force of the process, i.e., the concentration of the reagent  $c_R$  (Fig. 1) in mass transfer and of the heat-transfer coefficient  $\alpha$  on the temperature head  $\Delta T$  during boiling [1]. It can be seen from Fig. 1 that in mass transfer we find two regimes. In analogy with boiling we call the first regime developed bubble regime: with increasing  $c_R$ ,  $k$  also increases. In boiling, the second regime will be called film regime. In case of mass transfer the film regime cannot be effected because transport of the reagent across the gas film is impossible. This is one of the substantial differences of the processes under examination. However, the isolation by gas bubbles is so great that, as in boiling, the mass-transfer coefficient decreases with increasing  $c_R$ . In analogy with boiling, this process might be called the pseudofilm process. Finally, the maximum values of the mass-transfer coefficients correspond to the critical mass flows and make it possible to classify them.

Establishing quantitative dependences is a very difficult task. Since it is impossible to obtain an accurate solution of the problem of mass transfer with gas formation due to the lack of reliable information on separate acts of the process that would make it possible to formulate the problem in the form of a closed system of equations, the quantitative relations will be established with the aid of a system of generalized variables. These relations will be established separately for each regime.

In the range of developed bubble regime the principal part is played by the mixing of the liquid by gas bubbles. In accordance with the hydrodynamic theory of the diffusion boundary layer, its thickness can be estimated [3]

$$\delta \sim L / \sqrt[3]{Pr} \sqrt{Re}, \quad (1)$$

where  $L$  is the characteristic dimension of the body. In the case of dissolution of solids,  $L = d$  [3]. In criterial form, Eq. (1) is written as follows:

$$Nu \sim \sqrt[3]{Pr} \sqrt{Re}. \quad (2)$$

The exponent 0.5 of the Reynolds number corresponds to the conditions on the front of a sphere around which a liquid flows, the flow being laminar. Due to the separation of the boundary layer at the rear of the sphere and the turbulization of the flow, the exponent may increase to 0.9 [3]. The speed of the flow around the sphere, which finds expression in the Reynolds number, can be characterized through the rate of gasification [3]

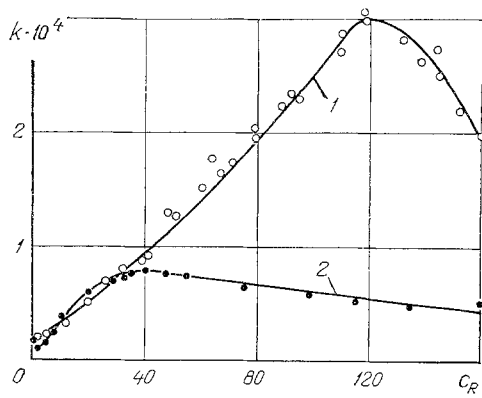


Fig. 1

Fig. 1. Dependence of the mass-transfer coefficient  $k$  (m/sec) on the concentration of the reagent  $c_R$  ( $\text{kg/m}^3$ ): 1) dissolution of magnesium in sulfuric acid; 2) dissolution of calcite in nitric acid.

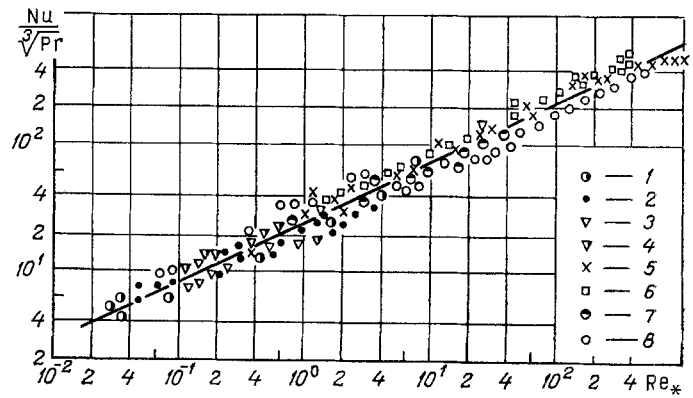


Fig. 2

Fig. 2. Experimental data on the kinetics of chemical boiling in the region of the bubble regime: 1)  $\text{CaCO}_3 + \text{HCl}$ ; 2)  $\text{CaCO}_3 + \text{HNO}_3$ ; 3)  $(\text{CuOH})_2\text{CO}_3 + \text{HCl}$ ; 4)  $(\text{CuOH})_2\text{CO}_3 + \text{HNO}_3$ ; 5)  $\text{Mg} + \text{HCl}$ ; 6)  $\text{Mg} + \text{H}_2\text{SO}_4$ ; 7)  $\text{Mn} + \text{HCl}$ ; 8)  $\text{Mg} + \text{HNO}_3$  (3, 4, 5 - data of [6]).

$$w \sim \frac{nk c_R}{\rho_g}, \quad (3)$$

and the Reynolds number is  $\text{Re}_* = nk c_R d / \rho_g v$ .

Processing of the experimental results and their representation in the form of the dependence  $\text{Nu} / \sqrt[3]{\text{Pr}} = f(\text{Re}_*)$  (Fig. 2) makes it possible to establish a theoretical relation for determining the intensity of mass transfer in the region of developed bubble regime

$$\text{Nu} = 24 \sqrt[3]{\text{Pr}} \sqrt{\text{Re}_*}. \quad (4)$$

It can be seen from Eq. (4) that the exponent is 0.5; this corresponds to the conditions for the existence of a laminar boundary layer.

In the region of the pseudofilm regime we must take into account, in addition to the factors of mixing, also the factors of insulation of the surface of the solid by gas bubbles. The proportion of the surface of mass transfer  $\psi$  occupied by the gas can be estimated from the product of the number of gasification centers  $N$  and the square of the separation diameter of the bubble  $d_0^2$ :

$$\psi \sim N d_0^2. \quad (5)$$

It follows from the analogy of the examined processes with heat exchange upon boiling that under conditions of constant pressure the number of gasification centers  $N$  is proportional to the difference of the gas concentrations whose magnitude is equal to  $nc_R$ . Bearing in mind that the results are represented in the form of generalized variables, we can estimate the magnitude of  $N$  from the diffusion Jacobi number  $\text{Ja}$  which, in the case of mass-transfer processes, is a measure of the ratio of the weight of the released gas as a result of the reaction in unit volume to the amount of gas that under the given conditions (temperature, pressure) can be contained in unit volume:  $\text{Ja} = nc_R / \rho_g$ .

Evaluating the results in determining the dependence of the number of gasification centers  $N$  on the Jacobi number  $\text{Ja}$  by the method of photography, we may assume that in the range of large Jacobi numbers this dependence is of a linear nature:  $N \sim \text{Ja}$ .

In regard to the separation diameter of the bubbles  $d_0$  the present author established jointly with Zelin'ski [2] by the method of photography that when  $\text{CO}_2$  is liberated (quasi-static conditions), its magnitude is constant and equal to  $7.5 \cdot 10^{-5}$  m. In the case of  $\text{H}_2$  being liberated (dynamic conditions), the separation diameter of the bubbles increases with increasing concentration of the reagent. For processes of heat exchange upon boiling under dynamic conditions, the separation diameter of the bubbles can be determined by the formula [4]

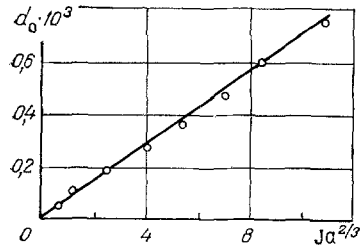


Fig. 3

Fig. 3. Dependence of the separation diameter of hydrogen bubbles  $d_0$  (m) on the value of  $Ja^{2/3}$ .

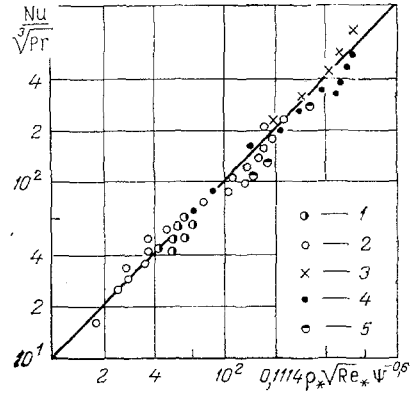


Fig. 4

Fig. 4. Comparison of the experimental data on the kinetic of chemical boiling in the region of the pseudofilm regime with the generalized dependence (9): 1)  $\text{CaCO}_3 + \text{HCl}$ ; 2)  $\text{CaCO}_3 + \text{HNO}_3$ ; 3)  $\text{Mg} + \text{HCl}$ ; 4)  $\text{Mg} + \text{H}_2\text{SO}_4$ ; 5)  $\text{Mn} + \text{HCl}$  (5 - data of [6]).

$$d_0 = 14.7 \sqrt[3]{\frac{\rho_l a^2}{g(\rho_l - \rho_g)}} Ja_g^{2/3}. \quad (6)$$

A similar dependence was obtained for the separation diameter of hydrogen bubbles

$$d_0 = 42.7 \sqrt[3]{\frac{\rho_R D_g^2}{g(\rho_R - \rho_g)}} Ja^{2/3}. \quad (7)$$

The experimental values of  $d_0$  in dependence on  $Ja^{2/3}$  upon liberation of hydrogen, presented in Fig. 3, confirm the analogous nature of the processes under examination. If we substitute into formula (7) the values of the physical magnitudes of the reagent and of hydrogen, then the separation diameter of the hydrogen bubbles is  $d_0 = 7.5 \cdot 10^{-5} Ja^{2/3}$ .

Since the number of gasification centers in formula (5) is evaluated by a dimensionless complex, the Jacobi number  $Ja$ , we represent the second term  $d_2^0$  in this product also in the form of a dimensionless simplex  $(d_0/d_x)^2$ , where  $d_x$  is the separation diameter of the hydrogen bubbles with the number  $Ja = 1$ . Such a diameter was chosen because with  $Ja = 1$  the separation diameter of the hydrogen bubbles is equal to the separation diameter of  $\text{CO}_2$  bubbles. Consequently, the value of  $\psi$  is estimated with an accuracy to the constant coefficient:

$$\psi \sim Ja \left( \frac{d_0}{d_x} \right)^2. \quad (8)$$

For  $\text{CO}_2$  bubbles  $\psi \sim Ja$  ( $d_0 = d_x$ ); for  $\text{H}_2$  bubbles  $\psi \sim Ja^{7/3}$ .

Finally, at places of intense heat or mass sinks we must take into account the existence of Archimedean buoyant forces. The calculation is most often carried out with the aid of a simplex representing the dimensionless density [5]  $\rho_* = \rho_l / \rho_g$ .

Taking the above-explained factors into account enabled us to generalize the experimental results in the region of the pseudofilm regime by the following criterial equation:

$$\frac{Nu}{\sqrt[3]{Pr}} = 0.1114 \rho_* \sqrt{Re_*} \psi^{-0.6}. \quad (9)$$

Figure 4 illustrates the comparison of the experimental values with dependence (9).

The critical values of the heat fluxes were determined in analogy to boiling in the form of a criterial correlation between the Archimedes and Reynolds numbers [5]. It was established from the experimental data that

$$Re_{* cr} = 3.8 \cdot 10^{-7} Ar^{0.7}. \quad (10)$$

The obtained quantitative dependences confirm the qualitative analogy of the processes of chemical boiling with heat exchange upon boiling.

#### NOTATION

$Nu = kd/D_R$ , Nusselt number;  $Re = wd/\nu$ , Reynolds number;  $Pr = \nu/D_R$ , Prandtl number;  $Ar = (gd^3/\nu^2)[(\rho_R - \rho_g)/\rho_g]$ , Archimedes number;  $Ja_H = (c_p \Delta T/r)(\rho_L/\rho_V)$ , heat Jacobi number;  $k$ , mass-transfer coefficient, m/sec;  $d$ , particle diameter, m;  $D$ , diffusion coefficient,  $m^2/sec$ ;  $\rho$ , density,  $kg/m^3$ ;  $\nu$ , viscosity of the liquid,  $m^2/sec$ ;  $g$ , acceleration of gravity,  $m/sec^2$ ;  $c_p$ , heat capacity with constant pressure,  $J/kg \cdot ^\circ K$ ;  $r$ , specific heat of vaporization,  $J/kg$ ;  $\alpha$ , thermal diffusivity,  $m^2/sec$ ;  $n$ , stoichiometric coefficient. Subscripts: R, reagent; g, gas; L, liquid; v, vapor.

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#### IMPREGNATION OF HEATED FILLER WITH VISCOPLASTIC LIQUID

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The article presents the calculation of the technological process of continuous impregnation of a heated porous layer (filler) moving in a viscoplastic liquid with the aid of the approximate parametric method of the boundary-layer theory.

The production of many composite materials includes the impregnation of porous fillers with various liquid binders which then, as a result of polymerization or crystallization, change into the solid state. Figure 1 shows one of the models of an impregnation apparatus dealt with in [1]. In chamber I the filler is heated in a gaseous medium to the temperature  $T_0$  (which may attain thousands of centigrades or more), and then it is drawn at constant speed  $u$  into chamber II filled with liquid binder at the temperature  $T_f < T_0$ . The pressure of the liquid  $p_f$  is greater than the air pressure  $p_0$  in the pores of the filler. Under the effect of the originating pressure gradient the liquid begins to enter the filler, thereby expelling the air from its pores, and at the distances  $x \geq L$  from the entrance into chamber II the liquid fills them completely. As a result of heat exchange the liquid is heated, its viscosity decreases, and the filtration rate increases.

From the solution of the problem of impregnation we have to determine in the first place the length  $L$  of the working section of the apparatus in dependence on the parameters of the process and the physical characteristics of the media, and a way of reducing this length while at the same time increasing the drawing speed; this is of interest in attempts to intensify the process under consideration. In the case of a filler having the form of a thin flat layer, and binder with the properties of an ordinary viscous liquid, this problem has a self-similar solution [1].

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