NOTATION

d, ρ and ε_0, diameter, density of the material, and porosity of the immobile filling of particles of the component; ε, porosity of the fluidized bed; q, thermal flux density; w, speed of filtration of the fluidizing medium; t, average temperature; ν and λ, coefficients of kinematic viscosity and thermal conductivity of the fluidizing medium; t_{sat}, saturation temperature; $D = 2\varepsilon d/3(1-\varepsilon)$, equivalent diameter of the pore channel; α, heat-transfer coefficient; $Nu = \alpha D/\lambda$, Nusselt criterion; $Re = wD/\varepsilon\nu$, Reynolds criterion; Pr, Prandtl criterion; ϑ, superheating of the heat-transfer surface; ϑ_0, same, in the case of developed surface boiling; Δq, effective thermal flux; m, a power index; $\Delta t = t_{sat} - t_{fl}$, underheating to the temperature of saturation; p, pressure; δ, diameter of the heater. Indices: fl, s, and fm, temperature of the fluidizing medium, the heat-transfer surface, and film.

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FILM BOILING WITH CHEMICAL REACTION ON A VERTICAL CATALYST SURFACE

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UDC 536.247

A mathematical model is given for the film boiling of a liquid on a vertical catalyst surface. The effect of the parameters of the mathematical description on the process is investigated.

There have been many works and review papers on film boiling on heated surfaces, reflecting the main features of the phenomenon [1, 2]. Exothermal chemical reaction on a catalyst surface between vapors of the vaporizing liquid leads to heating of the catalyst surface and film boiling. An important characteristic of this type of boiling is the dependence of the heat fluxes and the surface temperature both on the rate of the reaction and on the intensity of vaporization processes at the gas–liquid interface. It is of interest to write a mathematical description of laminar film boiling on the vertical surface of a catalyst plate of length L and thickness 2b.

The physical model of the process is shown in Fig. 1, where 2 is the liquid flowing past a vapor film 1 at some constant rate U_∞. As a result of heating of the metal-catalyst surface 3, the liquid vaporizes, and its vapors react on the active catalytic surface, maintaining its temperature at a level sufficient for stable film boiling. In deriving the mathematical description, it is assumed that mass and heat transfer in the vapor film in the longitudinal plate direction occurs as a result of the convective flux and mass and heat transfer in the transverse direction as a result of diffusion and heat conduction and also the transverse component of the vapor velocity.

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The heat-transfer equation for the catalyst plate in the longitudinal and transverse directions is

$$a\,\frac{\partial^2\Theta_3}{\partial H^2}+\frac{\partial^2\Theta_3}{\partial \xi^2}=0. \tag{1}$$

The continuity and momentum-transfer equations in the gas phase are

$$\frac{\partial \bar{U}}{\partial \xi}+\frac{\partial \bar{V}}{\partial H}=0, \tag{2}$$

$$\bar{U}\,\frac{\partial \bar{U}}{\partial \xi}+\bar{V}\,\frac{\partial \bar{U}}{\partial H}=\frac{\nu_1}{\nu_2}\cdot\frac{L^2}{R^2}\cdot\frac{1}{\mathrm{Re}}\cdot\frac{\partial^2 \bar{U}}{\partial H^2}+\frac{\Delta\rho}{\rho_1}\cdot\frac{1}{\mathrm{Fr}}. \tag{3}$$

The equations of mass and heat transfer through the moving gas film are

$$\frac{1}{\mathrm{Pe}_{1M}}\,\frac{\partial^2 C_1}{\partial H^2}=\bar{U}\,\frac{\partial C_1}{\partial \xi}+\bar{V}\,\frac{\partial C_1}{\partial H}, \tag{4}$$

$$\frac{1}{\mathrm{Pe}_{1T}}\,\frac{\partial^2 \Theta_1}{\partial H^2}=\bar{U}\,\frac{\partial \Theta_1}{\partial \xi}+\bar{V}\,\frac{\partial \Theta_1}{\partial H}. \tag{5}$$

Heat transfer in the liquid to the phase interface is described by the equation

$$\frac{1}{\mathrm{Pe}_2}\,\frac{\partial^2 \Theta_2}{\partial H^2}=\frac{\partial \Theta_2}{\partial \xi}. \tag{6}$$

Boundary conditions must be added to the system in Eqs. (1)-(6). For example, when

$$\xi=0,\ \Theta_3=\Theta_0;\quad \bar{U}=1;\ C_1=1;\ \delta=\delta_0, \tag{7}$$

$$\xi=1,\ \frac{\partial \Theta_3}{\partial \xi}=0, \tag{8}$$

$$H=-1,\ \frac{\partial \Theta_3}{\partial H}=0. \tag{9}$$

The boundary conditions at the catalyst surface correspond to conditions of the fourth kind, but with sources at the boundaries

$$\text{when } H=0,\ \frac{\partial \Theta_1}{\partial H}+\mathrm{DaLe}\,C_1\Delta\Theta_{\mathrm{ad}}\exp\left[\frac{\Theta}{1+\gamma\Theta_1}\right]=\frac{\lambda_1}{\lambda_3}\cdot\frac{\partial \Theta_3}{\partial H}, \tag{10}$$

$$\Theta_1=\Theta_3, \tag{11}$$

$$\frac{\partial C_1}{\partial H}=\mathrm{Da}\,C_1\exp\left[\frac{\Theta_1}{1+\gamma\Theta_1}\right], \tag{12}$$

$$\bar{U}=0,\quad \bar{V}=0; \tag{13}$$

$$\text{when } H=\delta(\xi),\ \bar{U}=1;\quad \Theta_1=0;\quad C_1=1;\quad \Theta_2=0; \tag{14}$$

$$\text{when } H\to\infty,\ \Theta_2=\Theta_0. \tag{15}$$

The second term in Eq. (10) and the right-hand side of Eq. (12) are determined by the conditions for a first-order chemical reaction at the catalyst surface:

$$\frac{\partial \Theta_1}{\partial H}-\frac{\lambda_2}{\lambda_1}\cdot\frac{\partial \Theta_2}{\partial H}=\frac{L}{b}\,\mathrm{Pe}_{1T}\,\mathrm{Ja}\,\frac{d}{d\xi}\int_0^{\delta}\bar{U}\,dH. \tag{16}$$

Equation (16) characterizes the heat-flux balance at the phase interface, and at the same time serves for the calculation of the gas-film thickness [3, 4].

The solution of the whole problem was found numerically on a Minsk-32 computer by the method of equilibration over time [5], for which the derivative $\partial\Theta_3/\partial t$ was added to Eq. (1). Then at each time step, the hydrodynamic equations were solved on the basis of the method proposed in [6], and hence, for a known velocity field, the concentration and temperature in the gas film and the catalyst grain were determined. The film thickness was calculated from Eq. (16) for the heat-flux values already determined. It was found that for a more accurate solution of the problem integration over the film thickness at each time step is expedient.

In the course of numerical analysis, the parameters of the system in Eqs. (1)-(16) were varied over the

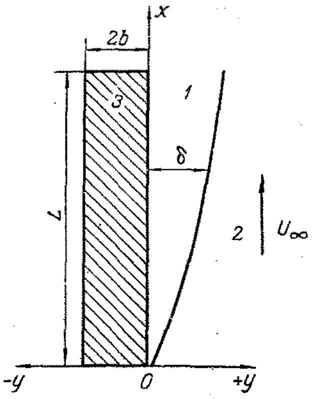

Fig. 1. Physical model of the process: 1) Vapor film; 2) liquid; 3) metal catalyst; U_∞, liquid velocity.

following ranges: Re = 100-1000; Fr = 100-1000; Pe_{1M} = 100-500; Pe_{1T} = 100-500; Ja = 0.1-5; Da = 1-5; Le = 0.1-1; $\Delta\Theta_{ad}$ = 1-5; b/L = 0.1-1; degree of underheating $\Theta_0 = -1-0$. Some of the results obtained are shown in Figs. 2-4.

For example, velocity (a), concentration (b), and temperature (c) profiles in the gas film and at the catalyst grain are shown in Fig. 2, where ξ is the longitudinal coordinate over the length of the plate; δ is the film thickness. For clarity, the coordinates for the determination of $\bar{U}$, C, Θ_1, and Θ_3 are shown for several values of ξ.

It is evident from the given results that the gas-flux velocity is a maximum at the phase interface, and then decreases linearly to zero; the concentration is a maximum at the phase boundary and linearly decreases with approach to the catalyst surface and with increase in the length ξ. The gas-phase temperature corresponds to saturation at the phase boundary, and then linearly increases with approach to the surface. There is little difference in catalyst temperature at the center of the plate and at the grain surface. The change in temperature over the length of the plate is more significant.

The change in surface temperature and heat fluxes depends on the intensity of heat liberation ($\Delta\Theta_{ad}$) (Fig. 3). It is evident from Fig. 3 that the heat flux is a maximum in the parts of the grain close to $\xi = 0$; the surface temperature of the catalyst gradually increases with increase in ξ until it reaches the adiabatic value, after which it remains constant.

The calculated values of the film thickness and mass fluxes over the length of the grain are shown in Fig. 4 as a function of the dimensionless constant Da. It follows from Fig. 4 that, at small Da, $\delta \sim \xi$, but with increase in reaction rate this relation becomes nonlinear and the film thickness comes to be proportional to $\sqrt{\xi}$. Note that in film conditions on the heating surface in nonflow conditions the film thickness is always proportional to $\xi^{1/4}$ [3]. As a result of numerical analysis it was established that the effect of Re on the temperature and concentration profiles is fairly slight: with increase in Re from 100 to 2000, δ decreases by ≈5-10%. The effect of the density ratio $\Delta\rho/\rho_1$ = 100-1000 and the ratio ν_2/ν_1 = 0.1-0.5 on the film thickness is neglibibly small. But the nature of the process and the numerical values of Θ, C, and δ are most affected by the parameters of the model characterizing the chemical reactions: Da, Le, $\Delta\Theta_{ad}$, and Ja.

On the basis of the numerical analysis, some simplifications may be made in the given mathematical model. For example, if

$$\varkappa = \frac{\nu_2}{\nu_1}\left(\frac{R}{L}\right)^2 \mathrm{Re} \ll 1, \tag{17}$$

$\varkappa$ is a small parameter, and the solution of the hydrodynamic part of the problem may be expanded in a series in powers of $\varkappa$. Then, confining attention to the zeroth approximation, it is found that

$$\bar{U}_0 = \frac{H}{\delta}. \tag{18}$$

As follows from Fig. 2, this is a fairly good approximation of the solution.

In view of the linearity of the concentration and temperature changes in the film, it may be assumed that

$$\frac{\partial\Theta_1}{\partial H} \approx \frac{\Theta_s}{\delta}; \quad \frac{\partial C_s}{\partial H} \approx \frac{1-C_s}{\delta}; \quad \frac{\partial\Theta_2}{\partial Z} = \Theta_0\sqrt{\frac{\mathrm{Pe}_2}{\pi\xi}}. \tag{19}$$

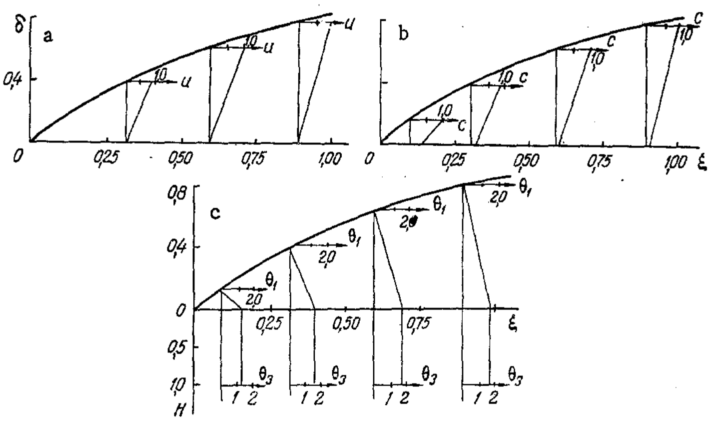

Fig. 2. Velocity (a), concentration (b), and temperature (c) profiles over the film thickness (δ) and the grain length (ξ). U, C, Θ_1, and Θ_3 are the ordinates of the corresponding quantities.

If the temperature drop between the surface of the grain and its center is slight (as follows from Fig. 2), $\partial\Theta_3/\partial H \approx 0$. Substitution of the results obtained in Eqs. (10) and (16) yields

$$\Theta_s = \frac{\lambda_2}{\lambda_1}\Theta_0\sqrt{\frac{\mathrm{Pe}_2}{\pi\xi}} + \frac{L}{2b}\mathrm{Pe}_{1T}\,\mathrm{Ja}\,\frac{d\delta}{d\xi}, \tag{20}$$

$$\Theta_s = \mathrm{Da}\,\mathrm{Le}\,\delta\Delta\Theta_{ad}\,\frac{\exp\left[\dfrac{\Theta_s}{1+\gamma\Theta_s}\right]}{1+\mathrm{Da}\,\delta\exp\left[\dfrac{\Theta_s}{1+\gamma\Theta_s}\right]}. \tag{21}$$

It follows from Eq. (21) that two extreme cases may be considered.

1. External-Diffusion Conditions

In this case, the heating of the catalyst is close to adiabatic, and

$$\frac{L}{2b}\mathrm{Pe}_{1T}\,\mathrm{Ja}\,\frac{d\delta}{d\xi} + \frac{\lambda_2}{\lambda_1}\Theta_0\sqrt{\frac{\mathrm{Pe}_2}{\pi\xi}} = \frac{\Delta\Theta_{ad}\mathrm{Le}}{\delta}. \tag{22}$$

Let $\delta = A\sqrt{\xi}$. It follows from Eq. (22) that

$$A = \frac{2b}{L}\cdot\frac{\lambda_2}{\lambda_1}\sqrt{\frac{\mathrm{Pe}_2}{\pi}}\cdot\frac{\Theta_0}{\mathrm{Pe}_{1T}\mathrm{Ja}}\left[-1+\sqrt{1+\frac{\pi}{4}\left(\frac{\lambda_1}{\lambda_2}\right)^2\frac{L}{b}\,\frac{\Delta\Theta_{ad}\mathrm{Le}}{\Theta_0^2}\,\frac{\mathrm{Pe}_{1T}\mathrm{Ja}}{\mathrm{Pe}_2}}\,\right]. \tag{23}$$

Thus, when the chemical reaction occurs in external-diffusion conditions, the film thickness varies in proportion to $\sqrt{\xi}$.

If the underheating $\Theta_0 = 0$, then

$$\delta = 2\sqrt{\frac{\Delta\Theta_{ad}\mathrm{Le}}{\mathrm{Pe}_{1T}\mathrm{Ja}}\cdot\frac{b}{L}\xi}. \tag{24}$$

2. Kinetic Conditions

For $\gamma \ll 1$ and small heating, $\exp[\Theta_s/(1+\gamma\Theta_s)] \approx 1+\Theta_s$. Then the equation for the calculation of the film thickness takes the form

$$\frac{L}{2b}\mathrm{Pe}_{1T}\mathrm{Ja}\,\frac{d\delta}{d\xi} + \frac{\lambda_2}{\lambda_1}\Theta_0\sqrt{\frac{\mathrm{Pe}_2}{\pi\xi}} = \mathrm{Da}\,\mathrm{Le}\,\Delta\Theta_{ad}, \tag{25}$$

from which the following result may be obtained:

$$\delta = \frac{2b}{L}\,\frac{\sqrt{\xi}}{\mathrm{Pe}_{1T}\mathrm{Ja}}\left[\mathrm{Da}\,\mathrm{Le}\,\Delta\Theta_{ad}\sqrt{\xi} - 2\frac{\lambda_2}{\lambda_1}\Theta_0\sqrt{\frac{\mathrm{Pe}_2}{\pi}}\right] + \delta_0. \tag{26}$$

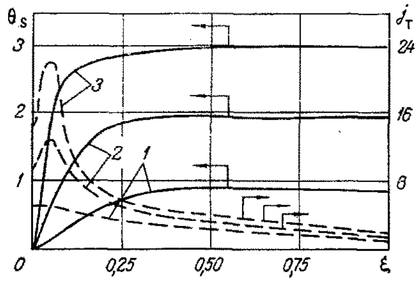

Fig. 3

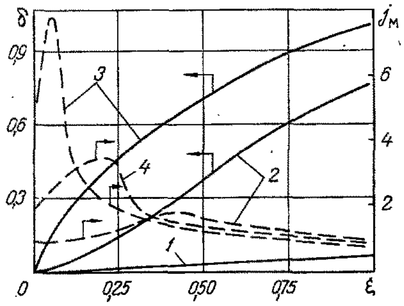

Fig. 4

Fig. 3. Change in surface temperature and heat fluxes at the grain surface as a function of the adiabatic heating: 1) $\Delta\Theta_{ad} = 1$; 2) 2; 3) 3. The parameter values are as follows: Da = 5; Le = 1; Re = 100; Fr = 100; b/L = 0.1; $\Theta_0 = 0$; $Pe_{1m} = 100$; Ja = 1.0. The continuous curves correspond to Θ_s and the dashed curves to the heat flux.

Fig. 4. Change in film thickness and mass fluxes over the grain length as a function of the chemical reaction rate: 1) Da = 0.1; 2) 1; 3) 5; 4) 2. The parameter values are as follows: $\Delta\Theta_{ad} = 3$; Re = 100; b/L = 0.1; Ja = 1.0. The continuous curves correspond to δ and the dashed curves to the mass fluxes.

If the underheating $\Theta_0 = 0$, then

$$\delta = \frac{2\,\mathrm{Da\,Le}\,\Delta\Theta_{ad}}{\mathrm{Pe}_{1T}\mathrm{Ja}} \cdot \frac{b}{L}\xi + \delta_0, \tag{27}$$

i.e., the film thickness is proportional to the length coordinate. Analogous results were obtained numerically in an analysis of the complete model (Figs. 2 and 3).

If the liquid is underheated to the boiling point, then, depending on the degree of underheating Θ_0, film conditions may only exist for certain values of the parameters.

Thus, integrating Eq. (26) over the length, it is found that this is the case when

$$\frac{3}{8}\,\frac{\mathrm{Da\,Le}\,\Delta\Theta_{ad}}{\frac{\lambda_2}{\lambda_1}\Theta_0}\sqrt{\frac{\pi}{\mathrm{Pe}_2}} > 1. \tag{28}$$

Numerical checking of the region of existence of film boiling in accordance with Eqs. (1)-(16) confirms that the condition in Eq. (28) is valid.

NOTATION

x and y, coordinates; L and b, length and half-thickness of plate; U_∞, velocity of unperturbed flux; ν, kinematic viscosity; D and a, diffusion coefficient and thermal diffusivity; λ, thermal conductivity; E, activation energy of reaction; $\bar{R}$, gas constant; T, temperature; C_p, specific heat; K, reaction rate constant; C, concentration; r_{vap}, heat of phase transformation; U and V, flux velocities; δ, film thickness; ρ, density; Q, heat output of reaction. Indices: 1, vapor; 2, liquid; 3, grain; 00, saturation; 0, reference value of the temperature, corresponding to the inflow point; y, x, directions of change in variables; T, M, thermal and mass parameters; s, grain surface. Dimensionless parameters: $\xi = x/L$; $H = y/b$; $\bar{U} = U_1/U_\infty$; $\bar{V} = (V/U_\infty)(L/b)$; $Re = U_\infty L/\nu_2$; $Fr = U_\infty^2/gL$; $Pe_{1m} = (U_\infty L/D_1)(b^2/L^2)$; $Pe_{1T} = (U_\infty L/a_1)(b^2/L^2)$; $Pe_2 = (U_\infty L/a_2)(b^2/L^2)$; $Da = K/D_1$; $Ja = (E/\bar{R}T_0^2)(r_{vap}\rho_1/C_{p_1})$; $Le = D_1/a_1$; $\Theta = [(T - T_0)/\bar{R}T_0^2]E$; $\gamma = \bar{R}T_0/E$; $\Delta\Theta_{ad} = (Q/C_{p1})\cdot(E/RT_0^2)$; $a = (\lambda_{3y}/\lambda_{3x})(L^2/b^2)$; $\Delta\rho/\rho_1 = (\rho_2 - \rho_1)/\rho_1$; $\Theta_0 = [(T_{00} - T_0)/\bar{R}T_0^2]E$; $Z = H - \delta$.

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HYDRODYNAMIC SITUATION AND TRANSFER PROCESSES IN THE STATIC GRANULAR BED OF A CATALYST

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Experimental data on the distribution of the local mass-transfer coefficients on the external surface of a spherical particle in a monodisperse static granular layer are considered. The observed rate of chemical change for a nonequiaccessible surface of the catalyst grains is determined.

It is shown in [1–2] that the free volume of a fixed layer of particles through which a gas or liquid flows is nonuniform – there is flowing region which is a jet, and a nonflowing region situated in the neighborhood of the points of contact between the particles. This hydrodynamic situation leads to nonequiaccessibility of the external surfaces of the particles, which has been experimentally studied in a number of papers [2–8]. In this paper we investigate the flow and transfer processes in a static granular layer and their effect of the rate of chemical change on the surface of the grains of catalyst. For this purpose we will use the experimental relations between the local Nusselt numbers on a grain in the static granular layer and the Reynolds number, which were electrochemically obtained [3], and we will compare them with the well-known theoretical relation

$$\mathrm{Nu} = A\,\mathrm{Re}^n,$$

corresponding to a certain structure of the flow and transfer mechanism. Experimental curves of $\mathrm{Nu} = A\mathrm{Re}^n$ for certain points on the surface of the particle are shown in Fig. 1. It can be seen from Fig. 1 that for points I ($\Theta = 22°$) the exponent $n = \tan\alpha = 0.5$, which corresponds to transfer processes in the laminar boundary layer in the region of running against the flow [9]. For points II ($\Theta = 135°$) the nature of this dependence changes: as the Reynolds number increases for $10 < \mathrm{Re} < 200$, $n = 0.5$, while for $200 < \mathrm{Re} < 2500$, $n = 0.65$, which corresponds to transfer processes in the laminar boundary layer in the region of separation for turbulent basic flow [9]. For points III ($\Theta = 157°$) up to $\mathrm{Re} \approx 250$ the local Nusselt number is independent of the Reynolds number, and it is then found to increase slightly. In this case $n \approx 0.3$, which corresponds to a viscous type of flow around this point, for which [10]

$$\mathrm{Re}_l = \frac{U_l d}{\nu} \ll 1.$$

The absence of a dependence of Nu on Re in the region $10 < \mathrm{Re} < 250$ indicates a molecular transfer mechanism at this point. It follows from this that the point considered is in the contact region between solid particles. However, as the Re number increases this region contracts, and the same point will lie in the viscous flow region.

For points IV ($\Theta = 225°$) the Nusselt number is independent of the Reynolds number over the whole range of numbers $10 < \mathrm{Re} < 2500$, i.e., the point IV is in the contact region.

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