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GAS FLOW IN A CAPILLARY WITH AN EXTERNAL DISTURBANCE VARYING THE COEFFICIENT OF MOLECULAR ADHESION

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UDC 532.63

The effect of change in the molecular adhesion coefficient on mass transfer in a capillary is studied for the free-molecular gas flow regime.

In recent years the interaction of laser radiation with material, leading to selective occurrence of many physicochemical reactions, has attracted ever greater interest of scientific investigators [1-3]. Among such reactions is a wide class of processes which occur on the boundary dividing two media. This class includes physical and chemical adsorption, heterogeneous catalysis, evaporation from the surface of solids and liquids, and diffusion in porous bodies. As was noted in [1], a correct understanding of the mechanisms by which they occur is extremely important for laser control of processes occurring on the surface of solids. Using the molecular-kinetic approach, the present study will consider certain questions related to the action of an external disturbance (in particular, laser radiation) on mass transfer in a capillary in the free-molecular gas flow regime.

As was noted in [1, 2] there are two possible variants of laser action on heterogeneous processes: 1) the radiation acts directly on the phase boundary and the molecules adsorbed thereon (the beam is incident on the surface); 2) the radiation acts upon the gas near the surface (the beam is parallel to the surface).

In the first case, aside from such possibilities as desorption stimulated by radiation and surface mobility of adsorbed molecules, one must also consider the possibility of direct radiation action on the adsorbent (producing changes in its catalytic properties, heating, etc.). Since clarification of all possible details of laser action on the material is quite difficult, in the first approximation we should select some parameters which

A. V. Lykov Institute of Heat and Mass Transfer, Academy of Sciences of the Belorussian SSR, Minsk. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 38, No. 3, pp. 507-513, March, 1980. Original article submitted June 15, 1979. will consider this action as a whole. Thus, in the first type of interaction one of these parameters could probably be the mean duration of a molecule's stay upon the surface (the adsorption time).

Below we will consider the second possible type of laser action on heterogeneous processes. In this case the basis of selective action upon transfer processes is the difference in the interaction with the surface of excited and unexcited molecules.

Falling upon the surface, a molecule may either be elastically reflected, or adhere to the surface (be adsorbed), with the probability of reflection being dependent on the net energy of the molecule E, which in the general case will include nonequilibrium oscillatory excitation energy which may be realized by laser action [1, 2].

Following [1], for the adhesion coefficient  $\alpha$  we have the expression

$$\alpha = 1 - \beta = 1 - \exp\left\{-\frac{E_s}{E}\right\},\tag{1}$$

where  $\beta$  is the reflection coefficient and  $E_s$  is the specific energy of molecular interaction with the wall.

We note that according to Eq. (1) with fulfillment of the condition  $E_S \gg E$  the adhesion coefficient is close to unity. However when this inequality is not fulfilled, one must consider the possibility of elastic reflection of molecules.

The division into two classes of laser interaction on heterogeneous processes introduced above is, of course, arbitrary. A laser beam incident on the surface can naturally also excite molecules in the gas phase. On the other hand, in the second type of interaction heat liberation may occur because of deactivation of excited molecules on the surface [4, 5], with consequent change in the adsorption time and surface diffusion coefficient. However, for a better understanding of the mass transfer process mechanism it is desirable in the first approximation to consider the individual effects of the various factors. Mass transfer in a capillary with varying adsorption times was considered in [6]. Here we will consider the effect of change in the adhesion coefficient, which characterizes the interaction of gas molecules with the surface, upon gas flow in a capillary.

We will consider the following model problem. Let the laser radiation act upon the gas near one end of a capillary of length L and radius r (parallel to the capillary end face). We assume that the laser beam passes so close to the end surface that all molecules entering the capillary from this end (X = L) fall within the zone of radiation action. We will assume that the molecules which have entered the capillary from the laser action zone are characterized by an adhesion coefficient  $\alpha_1$ . The molecules which enter the capillary at X = 0 will be assumed to be unexcited, with an adhesion coefficient  $\alpha_2$ . Further, we assume that the molecular oscillatory relaxation time of molecules adsorbed on the surface is much less than the mean adsorption time, so that molecules desorbed from the surface are also unexcited.

It is well known that in sufficiently thin capillaries mass transfer is accomplished both in the gas phase and by surface diffusion [7]. Naturally, only molecules adhering to the surface participate in surface diffusion (i.e., only those molecules which adhere to the surface for some time), while molecules which are reflected elastically from the surface are transferred into the gas phase. We will consider a system characterized by a constant adsorption time  $\tau$ , surface diffusion coefficient D<sub>S</sub>, and identical values of molecular flux entering the capillary at X = 0 and X = L. In the absence of a source of excitation for oscillatory degrees of freedom of the gas molecules, there will be no mass transfer through the capillary (the density of adsorbed molecules will be the same everywhere). Upon action of a laser which changes the adhesion coefficient of molecules near one end of the capillary (X = L), the density of adsorbed molecules near X = L changes, which leads to surface diffusion of molecules. Equilibrium in the system is disrupted and a resultant mass flux through the capillary will be observed.

In deriving the equations for the degree of coating of the capillary's interior surface by adsorbed molecules  $\Theta$ , we will make use of the premise of Langmuir's monomolecular adsorption theory that molecules falling on a portion of the surface already occupied by adsorbed molecules will be reflected back into the gas phase. However, in contrast to the usual assumption that all molecules incident on spots on the surface free of adsorbed molecules will be adsorbed, here we will consider the possibility of their reflection, as outlined above.

We write the continuity equation for an element of the adsorption layer in the stationary case

$$-\operatorname{div} J_s = J_d - \alpha_i (1 - \Theta) J^-,$$

(2)

where  $J_s$  is the density of the molecular flux transferred along the surface in the adsorption layer;  $J_d$ , flux of molecules desorbed per unit channel length;  $J^-$ , flux of molecules incident upon a unit surface. The quantity  $\alpha_i$  in Eq. (2), according to Eq. (1), is dependent on whether the molecules fall on the surface in excited or unexcited states. The quantities  $J_s$ ,  $J_d$  have the form:

$$J_s = -D_s n_0 \frac{d\Theta}{dX} , \quad J_d = \frac{n_0 \Theta}{\tau} . \tag{3}$$

For a cylindrical channel the flux density of departing molecules  $J^+$ , which includes desorbed  $(J_d)$  and reflected (J) molecules, is related to the flux of incident molecules  $J^-$  in the following manner (all departing molecules are assumed distributed over a cosine law) [8]:

$$J^{-}(x) = \int_{0}^{1} J^{+}(x') K_{1}(|x-x'|) dx' + N_{0}K(x) + N_{1}K(1-x), \qquad (4)$$

where x = X/L; N<sub>0</sub>, N<sub>1</sub> are the flux densities of molecules entering the capillary at x = 0 and x = 1, which we will assume equal to each other in the future; K<sub>1</sub>, K are functions characterizing the probability of movement of molecules from one surface element to another.

With the aid of Eqs. (2) - (4), to determine the degree of coating of the surface by adsorbed molecules we obtain a system of equations

$$\frac{n_{0}}{L^{\circ}} \frac{d}{dx} \left( D_{s} \frac{d\Theta}{dx} \right) = \frac{n_{0}\Theta}{\tau} - (1 - \Theta) \alpha_{2} \left[ \int_{0}^{1} \left( J_{2} + \frac{n_{0}\Theta}{\tau} \right) K_{1} \left( |x - x'| \right) dx' + N_{0}K(x) \right] - (1 - \Theta) \alpha_{1} \left[ \int_{0}^{1} J_{1}K_{1} \left( |x - x'| \right) dx' + N_{0}K(1 - x) \right];$$
(5)

$$J_{i} = [1 - \alpha_{i} (1 - \Theta)] \left[ \int_{0}^{1} J_{i} K_{i} (|x - x'|) dx' + N_{0} K (1 - x) \right];$$
(6)

$$J_{2} = [1 - \alpha_{2} (1 - \Theta)] \left[ \int_{0}^{1} \left( J_{2} + \frac{n_{0}\Theta}{\tau} \right) K_{i} (|x - x'|) dx' + N_{0} K(x) \right].$$
(7)

Here  $J_1$ ,  $J_2$  are the flux densities of elastically reflected molecules in excited and unexcited states, respectively.

Following [9], on the end surfaces we assign the conditions

$$\frac{d\Theta}{dx}\Big|_{x=0} = \frac{L}{\sqrt{D_s\tau}} \ [\Theta(0) - \Theta_{0e}]; \tag{8}$$

$$\frac{d\Theta}{dx}\Big|_{x=1} = \frac{L}{\sqrt{D_s\tau}} \quad [\Theta_{ie} - \Theta(1)], \tag{9}$$

where

$$\Theta_{0e} = \frac{\alpha_2 N_0 \tau}{n_0 + \alpha_2 N_0 \tau} ; \quad \Theta_{1e} = \frac{\alpha_1 N_1 \tau}{n_0 + \alpha_1 N_1 \tau} .$$

The total molecular flux  $N_r$  departing from the capillary through the section x = 1 consists of molecules which have passed through the capillary without colliding with the wall, molecules which pass through the section x = 1 after desorption or elastic reflection from the wall, and a surface molecular flux. Thus for  $N_r$  we have the expression

$$N_{r} = \pi r^{2} \left[ N_{0} K_{2} (1) + 2l \int_{0}^{1} \left( \frac{n_{0} \Theta}{\tau} + J_{1} + J_{2} \right) K (1-x) dx \right] - \frac{2\pi r}{L} D_{s} n_{0} \left. \frac{d\Theta}{dx} \right|_{x=1}.$$
 (10)

Now, for simplicity we will analyze the solution of Eqs. (5)-(9) for  $\Theta \ll 1$  (fulfillment of Henry's law), and also assume that the adhesion coefficient of unexcited molecules  $\alpha_2$  is equal to unity. In this case Eqs. (5)-(9) may be rewritten as:

$$\frac{D_{\mathbf{s}}\tau}{L^2} \frac{d^2n}{dx^2} = n - \int_0^1 nK_1 \left( |x - x'| \right) \, dx' - \alpha_1 \tau \int_0^1 J_1 K_1 \left( |x - x'| \right) \, dx' - \tau N_0 K \left( x \right) - \alpha_1 \tau N_0 K \left( 1 - x \right); \tag{11}$$

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$$J_{1} = (1 - \alpha_{1}) \left[ \int_{0}^{1} J_{1}K_{1} \left( |x - x'| \right) dx' + N_{0}K \left( 1 - x \right) \right];$$
(12)

$$\frac{dn}{dx}\Big|_{x=0} = \frac{L}{\sqrt{D_s\tau}} [n(0) - N_0\tau], \qquad (13)$$

$$\frac{dn}{dx}\Big|_{x=1} = \frac{L}{\sqrt{D_s\tau}} \quad [\alpha_1 N_0 \tau - n (1)].$$
(14)

In solving system (11)-(14) and calculating the resultant flux (10) we will approximate functions K,  $K_1$ ,  $K_2$  by exponential expressions [10]. The solution of Eq. (12) then has the form

$$J_1 = c_1 \exp\left\{-l \sqrt{\alpha_1} x\right\} + c_2 \exp\left\{l \sqrt{\alpha_1} x\right\},\tag{15}$$

where

$$c_{1} = \frac{(1+\sqrt{\alpha_{1}})(\sqrt{\alpha_{1}}-1)^{2}N_{0}}{(\sqrt{\alpha_{1}}-1)^{2}\exp\left\{-l\sqrt{\alpha_{1}}\right\}-(1+\sqrt{\alpha_{1}})^{2}\exp\left\{l\sqrt{\alpha_{1}}\right\}};$$
  

$$c_{2} = \frac{(1+\sqrt{\alpha_{1}})^{2}(\sqrt{\alpha_{1}}-1)N_{0}}{(\sqrt{\alpha_{1}}-1)^{2}\exp\left\{-l\sqrt{\alpha_{1}}\right\}-(1+\sqrt{\alpha_{1}})^{2}\exp\left\{l\sqrt{\alpha_{1}}\right\}}.$$

From Eqs. (11), (15), as in [9], for n we obtain the following differential equation:

$$\frac{d^4n}{dx^4} - \lambda \frac{d^2n}{dx^2} = \varphi \exp\left\{l\sqrt{\alpha_1}x\right\} - \psi \exp\{-l\sqrt{\alpha_1}x\},\tag{16}$$

where

$$\lambda = l^2 + \frac{1}{G}$$
;  $G = \frac{\tau D_s}{L^2}$ ;  $\varphi = \frac{\alpha_1 \tau c_2 l^2}{G}$ ;  $\psi = -\frac{\alpha_1 \tau c_1 l^2}{G}$ .

From Eq. (16) we have

$$n = \frac{a_1}{\lambda} \exp\left\{\sqrt{\lambda}x\right\} + \frac{a_2}{\lambda} \exp\left\{-\sqrt{\lambda}x\right\} + \frac{b_1}{l^2\alpha_1} \exp\left\{l\sqrt{\alpha_1}x\right\} + \frac{b_2}{l^2\alpha_1} \exp\left\{-l\sqrt{\alpha_1}x\right\} + g_1x + g_2, \tag{17}$$

where

$$b_1 = rac{\Phi}{l^2 lpha_1 - \lambda}$$
;  $b_2 = -rac{\psi}{l^2 lpha_1 - \lambda}$ ,

while for  $a_1$ ,  $a_2$ ,  $g_1$ ,  $g_2$ , upon substituting Eq. (17) in the original equation (11) we obtain the following system of algebraic equations:

$$\begin{aligned} -\frac{a_1}{\lambda \left(\sqrt{\lambda}+l\right)} + \frac{a_2}{\lambda \left(l-\sqrt{\lambda}\right)} - \frac{g_1}{l^2} + \frac{g_2}{l} &= \frac{2\sigma}{l} - \frac{b_1}{l^3 \alpha_1 \left(\sqrt{\alpha_1}+1\right)} - \frac{b_2}{l^3 \alpha_1 \left(1-\sqrt{\alpha_1}\right)}; \\ \frac{a_1}{\lambda \left(\sqrt{\lambda}-l\right)} &\exp\left\{\sqrt{\lambda}-l\right\} - \frac{a_2}{\lambda \left(\sqrt{\lambda}+l\right)} &\exp\left\{-\left(\sqrt{\lambda}+l\right)\right\} - \\ -g_1\left(\frac{1}{l} + \frac{1}{l^2}\right) &\exp\left\{-l\right\} - \frac{g_2}{l} &\exp\left\{-l\right\} = \frac{2\nu}{l} - \\ -\frac{b_1}{l^3 \alpha_1 \left(\sqrt{\alpha_1}-1\right)} &\exp\left\{l \left(\sqrt{\alpha_1}-1\right)\right\} + \frac{b_2}{l^3 \alpha_1 \left(\sqrt{\alpha_1}+1\right)} &\exp\left\{-l \left(\sqrt{\alpha_1}+1\right)\right\}; \\ a_1\left(\frac{1}{\sqrt{\lambda}} - \frac{1}{\lambda \sqrt{G_L}}\right) - a_2\left(\frac{1}{\sqrt{\lambda}} + \frac{1}{\sqrt{G_L}\lambda}\right) + g_1 - \frac{g_2}{\sqrt{G_L}} = \\ &= \frac{1}{\sqrt{G_L}} \left(\frac{b_1}{l^2 \alpha_1} + \frac{b_2}{l^2 \alpha_1} - \tau N_0\right) - \frac{b_1}{l \sqrt{\alpha_1}} + \frac{b_2}{l \sqrt{\alpha_1}}; \\ a_1\left(\frac{1}{\sqrt{\lambda}} + \frac{1}{\sqrt{G_L}\lambda}\right) \exp\left\{\sqrt{\lambda}\right\} - a_2\left(\frac{1}{\sqrt{\lambda}} - \frac{1}{\sqrt{G_L}\lambda}\right) \exp\left\{-\sqrt{\lambda}\right\} + \\ &+ g_1\left(1 + \frac{1}{\sqrt{G_L}}\right) + \frac{1}{\sqrt{G_L}} g_2 = \frac{b_2}{l \sqrt{\alpha_1}} \exp\left\{-l \sqrt{\alpha_1}\right\} - \end{aligned}$$

TABLE 1. Quantity  $\delta$  versus Capillary Radius (L =  $10^{-5}$  cm, D<sub>s</sub> =  $10^{-5}$  cm<sup>2</sup>/sec,  $\tau = 10^{-5}$  sec,  $\alpha_1 = 0.8$ )

r, cm	į0−e	2·10 <sup>-6</sup>	3.10-6	4.10-6	5.10-6	6·10 <sup>-6</sup>	7.10-6	8.10-6	9·10 <sup>-6</sup>	10-5
δ	1,334	0,669	0,447	0,335	0,267	0,223	0,191	0,167	0,148	0,133

TABLE 2. Quantity  $\delta$  versus Adsorption Time (L =  $10^{-5}$  cm, D<sub>S</sub> =  $10^{-5}$  cm<sup>2</sup>/sec, r =  $10^{-6}$  cm,  $\alpha_1 = 0.8$ )

τ, sec	10-6	2.10-6	3.10-6	4.10-6	5.10-6	6 • 10 <sup>-6</sup>	7·10 <sup>-6</sup>	8.10-6	9.10-6	10-5
δ	0,24	0,417	0,568	0,703	0,826	0,940	1,046	1,147	1,243	1,334

TABLE 3. Quantity  $\delta$  versus Adhesion Coefficient (L =  $10^{-5}$  cm, D<sub>S</sub> =  $10^{-5}$  cm<sup>2</sup>/sec, r =  $10^{-6}$  cm,  $\tau = 10^{-5}$  sec)

α1	0,55	0,6	0,65	0,7	0,75	0,8	0,85	0,9	0,95	1
δ	3,008	2,673	2,337	2,003	1,668	1,334	1	0,667	0,333	0

$$-\frac{b_1}{l\sqrt{\alpha_1}}\exp\{l\sqrt{\alpha_1}\}+\frac{1}{\sqrt{G_L}}\left(N_0\alpha_1\tau-\frac{b_1}{l^2\alpha_1}\exp\{l\sqrt{\alpha_1}\}-\frac{b_2}{l^2\alpha_1}\exp\{-l\sqrt{\alpha_1}\}\right)$$

where

$$\sigma = \frac{N_0 \tau}{2} - \frac{\alpha_1 \tau}{2} \left( \frac{c_1}{1 - \sqrt{\alpha_1}} + \frac{c_2}{1 + \sqrt{\alpha_1}} \right);$$
  
$$v = \frac{\alpha_1 \tau}{2} \left[ \frac{c_1}{\sqrt{\alpha_1} + 1} \exp\left\{-l\left(\sqrt{\alpha_1} + 1\right)\right\} - \frac{c_2}{\sqrt{\alpha_1} - 1} \exp\left\{l\left(\sqrt{\alpha_1} - 1\right)\right\} - \frac{N_0 \alpha_1 \tau}{2} \exp\left\{-l\right\}.$$

The flux  $N_r$  is calculated with Eq. (10). Tables 1-3 present values of the quantity  $\delta = (N_r - \pi r^2 N_0)/\pi r^2 N_0$ , which determines the ratio of the flux of molecules exiting the capillary to the flux of molecules entering the capillary, and characterizes the effect of change in the adhesion coefficient on mass transfer in the capillary.

As is evident from the tables,  $\delta$  decreases with increase in r and  $\alpha_1$  and increases with increase in  $\tau$ . In the case of a flow of a mixture of gases through the capillary, where under the conditions specified above the mass transfer of the individual components within the capillary is independent, with the aid of  $\delta_i$  for the individual components we can characterize the effect of mixture separation at the capillary output. Since in principle the value  $\delta_i$  depends on which component is excited by the laser radiation, we have, in principle, the possibility of selective separation of a gas mixture.

In conclusion, we will note that heating of the gas within the volume adjacent to the capillary at x = 1 due to redistribution of energy between oscillatory and translational degrees of freedom produced by intermolecular collision can be neglected given a certain ratio between the gas density and the characteristic dimensions of the system. In particular, [11] presents an estimate of the parameter range over which relaxation of the oscillatorily excited gas near the wall can be no more significant than the usual relaxation connected with paired collisions.

### NOTATION

n, density of adsorbed molecules;  $n_0$ , density of molecules in completely coated monolayer;  $\Theta = n/n_0$ , degree of surface coating by adsorbed molecules; l = L/r.

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## EFFECT OF A COOLANT TEMPERATURE

# JUMP ON A CLAD FUEL ROD

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An analytic expression is derived for the time dependence of the temperature distribution in cylindrical clad fuel elements. The transient state results from a jump in the coolant temperature.

We assume that the coolant temperature in a nuclear reactor operating in a steady state is instantaneously increased from  $T_F$  to  $T_F + \Delta T$ . Such an increase can arise because of leakage of core coolant, as the result of an accident, or for other reasons. It is assumed that the heat release (in fission) rate remains unchanged, and consequently the temperature of the fuel and cladding increases until a new steady state is reached. The solution of the transient problem resulting from such a hypothetical accident is important for two reasons. First, it is expedient and necessary to know a priori whether the temperature of the fuel or cladding at the end of the transient process reaches dangerous values which imperil the effectiveness and safety of the operation of the facility [1]. Second, it is important to establish from the variation of temperature with time whether the material, which was already subjected to a heat load because of the spatial temperature gradient, experiences further heat loads as a consequence of the accidental jump in temperature. Heat loads are particularly dangerous in a transient process.

Thus, the present problem is reduced to an emergency situation which can occur in a nuclear reactor, and its solution would permit an appropriate choice of materials and operating conditions of the facility. We assume that the fuel element and cladding are homogeneous and isotropic and have constant physical characteristics. In cylindrical coordinates the heat-conduction equations for a fuel element and cladding are, respectively,

$$\frac{\alpha_{i}}{\alpha_{2}}\left[\frac{\partial^{2}\tilde{\theta}_{i}}{\partial\xi^{2}}+\frac{1}{\xi}\frac{\partial\tilde{\theta}_{i}}{\partial\xi}+\frac{\partial^{2}\tilde{\theta}_{i}}{\partial\zeta^{2}}+q_{\theta}\cos\pi b\zeta\right]=\frac{\partial\tilde{\theta}_{i}}{\partial\operatorname{Fo}},\qquad(1)$$

$$\frac{\partial^2 \tilde{\theta}_2}{\partial \xi^2} + \frac{1}{\xi} \frac{\partial \tilde{\theta}_2}{\partial \xi} + \frac{\partial^2 \tilde{\theta}_2}{\partial \xi^2} = \frac{\partial \tilde{\theta}_2}{\partial F_0} . \tag{2}$$

The functions  $\tilde{\theta}_1$  and  $\tilde{\theta}_2$  and the parameters in Eqs. (1) and (2) are defined as follows:

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