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NONLINEAR MASS TRANSFER BETWEEN A GAS AND A FALLING LIQUID FILM.

3. MULTICOMPONENT MASS TRANSPORT

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UDC 532.72:532.529

A solution is obtained for the problem of multicomponent mass transfer between a gas and a falling liquid film. The case is considered in which the mass transfer of one of the components is limited by the nonlinear mass transport in the gas phase. The rates of multicomponent mass transport in the gas and liquid phases are determined.

In the first and second parts of this work [1, 2] it was shown that nonlinear mass transfer (as a result of intensive mass transport in the gas phase) leads to significant changes in the velocity distributions in the liquid and gas. In the case of multicomponent mass transport this leads not only to changes in the rates of transport of the components with the large concentration gradients but also to changes in the rates of transport of the components for which the concentration gradients do not influence the hydrodynamics of the flow.

The literature contains a number of experimental studies [3-5] in which it has been shown that as a result of simultaneous mass transfer in gas-liquid and liquid-liquid systems the transport of one component leads to changes in the rates of transport of the others. In these cases an increase in the rate of mass transfer is usually observed which is caused by the Marangoni effect, since this cannot be explained using the linear theory of mass transport [6].

In the present paper the effect will be considered for the case of multicomponent mass transport for the case in which the concentration gradient of one of the components in the gas phase influences the hydrodynamics of the flow.

Mathematical Description. The theory of diffusion in multicomponent systems [7, 8] shows that the approximation of independent diffusion can be used not only in the case when the concentrations of the components are low, but also when the diffusion coefficients of the individual components are similar. This makes it possible to solve the problem of the kinetics of nonlinear mass transfer between a gas and a falling liquid film in multicomponent systems by solving the problem for the transport of any component to the approximation of the linear theory of mass transport, where the velocity distribution takes into account the effects of the nonlinear mass transport of one of the components.

Let $\bar{c}_1$ and c_1 be the concentrations of any component in the gas and liquid, where mass

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Institute of Chemical Engineering, Bulgarian Academy of Sciences, Sofia. Translated from Inzhenerno-fizicheskii Zhurnal, Vol. 59, No. 4, pp. 593-602, October, 1990. Original article submitted January 23, 1989.

0022-0841/90/5904-1272$12.50

transport of this component does not influence the hydrodynamics of the flow. The distributions of these concentrations are determined from the system of equations

$$\begin{gathered}\tilde u \frac{\partial \tilde c_1}{\partial x} + \tilde v \frac{\partial \tilde c_1}{\partial y} = \tilde D_1 \frac{\partial^2 \tilde c_1}{\partial y^2};\\ x = 0,\ \tilde c_1 = \tilde c_{10};\quad y = h,\ \tilde c_1 = \chi_1 c_1;\quad y \to \infty,\ \tilde c_1 = \tilde c_{10};\\ u \frac{\partial c_1}{\partial x} + v \frac{\partial c_1}{\partial y} = D_1 \frac{\partial^2 c_1}{\partial y^2};\\ x = 0,\ c_1 = c_{10};\quad y = 0,\ \frac{\partial c_1}{\partial y} = 0;\quad y = h,\ I_1 = \tilde I_1,\end{gathered} \tag{1}$$

where the mass fluxes I_1 and $\bar I_1$ have diffusional and convective components as a result of the nonlinear mass transport of a component with a large concentration gradient:

$$\begin{gathered}\tilde I_1 = -M_1 \tilde D_1 \left(\frac{\partial \tilde c_1}{\partial y}\right)_{y=h} + M_1 \tilde v_n (\tilde c_1)_{y=h},\\ I_1 = -M_1 D_1 \left(\frac{\partial c_1}{\partial y}\right)_{y=h} + M_1 v_n (c_1)_{y=h},\\ \tilde v_n = -\frac{M\tilde D}{\tilde\rho_0^*}\left(\frac{\partial \tilde c}{\partial y}\right)_{y=h},\quad v_n = -\frac{MD}{\rho_0^*}\left(\frac{\partial c}{\partial y}\right)_{y=h}.\end{gathered} \tag{2}$$

In Eqs. (1), (2) the distributions of the velocities and concentrations are determined in [1, 2] taking into account the nonlinear mass transport of one of the components in the gas phase. As in [1, 2], use is made here of a mathematical description to the zeroth approximation with respect to the small parameter $(h_0/\ell)^2$.

The solution of the problem (1) makes it possible to determine the mass transfer rate:

$$J_1 = M_1 \tilde k_1 (\tilde c_{10} - \chi_1 c_{10}) = \frac{1}{l}\int_0^l \tilde I_1 dx = M_1 k_1 (\tilde c_{10}/\chi_1 - c_{10}) = \frac{1}{l}\int_0^l I_1 dx.$$

The Sherwood numbers for the gas and the liquid are found from Eq. (2):

$$\begin{gathered}\tilde{\mathrm{Sh}}_1 = \frac{\tilde k_1 l}{\tilde D_1} = \frac{1}{M_1 \tilde D_1 (\tilde c_{10} - \chi_1 c_{10})}\int_0^l \tilde I_1 dx,\\ \mathrm{Sh}_1 = \frac{k_1 l}{D_1} = \frac{\chi_1}{M_1 D_1 (\tilde c_{10} - \chi_1 c_{10})}\int_0^l I_1 dx.\end{gathered} \tag{3}$$

On converting in Eq. (1) to the dimensionless variables

$$\begin{gathered}X = \frac{x}{l},\ \tilde Y = \frac{y - h}{\tilde\delta},\ \tilde U(X,\ \tilde Y) = \frac{\tilde u}{\tilde u_0},\ \tilde V(X,\ \tilde Y) = \frac{\tilde v}{\tilde\varepsilon_0 \tilde u_0},\\ H(X) = \frac{h}{h_0},\quad Y = \frac{y}{h_0},\ U(X,\ Y) = \frac{u}{u_0},\quad V(X,\ Y) = \frac{v}{\varepsilon_0 u_0},\\ \tilde C_1 = \frac{\tilde c_1 - \chi_1 c_{10}}{\tilde c_{10} - \chi_1 c_{10}},\quad C = \frac{c_1 - c_{10}}{\tilde c_{10}/\chi_1 - c_{10}},\end{gathered} \tag{4}$$

where

$$\varepsilon_0 = \frac{h_0}{l};\ \tilde\varepsilon_0 = \frac{\tilde\delta}{l};\quad u_0 = \frac{g h_0^2}{3\nu};\ \tilde\delta = \sqrt{\frac{\tilde D l}{\tilde u_0}}.$$

the problem (1) assumes the following form in terms of the new variables

$$\tilde U \frac{\partial \tilde C_1}{\partial X} + \tilde V \frac{\partial \tilde C_1}{\partial \tilde Y} = \left(\frac{\varepsilon}{\varepsilon_1}\right)^2 \frac{\partial^2 \tilde C_1}{\partial \tilde Y^2} + \delta_0 H' \tilde U \frac{\partial \tilde C_1}{\partial \tilde Y}; \tag{5}$$

$$
\begin{gathered}
X=0,\ \tilde{C}_1=1;\\
\tilde{Y}=0,\ \tilde{C}_1=(C_1)_{Y=H};\\
\tilde{Y}\to\infty,\ \tilde{C}_1=1;\\
U\frac{\partial C_1}{\partial X}+V\frac{\partial C_1}{\partial Y}=\mathrm{Fo}_1\frac{\partial^2 C_1}{\partial Y^2};\\
X=0,\quad C_1=0;\\
Y=0,\ \frac{\partial C_1}{\partial Y}=0;\\
Y=H,\ \frac{\partial C_1}{\partial Y}=\beta_1\left(\frac{\partial \tilde{C}_1}{\partial \tilde{Y}}\right)_{\tilde{Y}=0}+\\
+\theta_3\beta_1\frac{\tilde{D}}{\tilde{D}_1}\left[\frac{\chi_1 c_{10}}{\tilde{c}_{10}-\chi_1 c_{10}}+(\tilde{C}_1)_{\tilde{Y}=0}\right]\left(\frac{\partial \tilde{C}}{\partial \tilde{Y}}\right)_{\tilde{Y}=0}-\\
-\varepsilon_0\theta_3\frac{\tilde{D}\tilde{\rho}^*}{D_1\rho_0^*}\left[\frac{\chi_1 c_{10}}{\tilde{c}_{10}-\chi_1 c_{10}}+(C_1)_{Y=H}\right]\left(\frac{\partial \tilde{C}}{\partial \tilde{Y}}\right)_{\tilde{Y}=0},
\end{gathered}
\tag{6}
$$

where

$$
\varepsilon_1=\tilde{\mathrm{Sc}}_1^{1/2};\ \varepsilon=\tilde{\mathrm{Sc}}^{1/2};\ \tilde{\mathrm{Sc}}_1=\frac{\tilde{\nu}}{\tilde{D}_1};\ \tilde{\mathrm{Sc}}=\frac{\tilde{\nu}}{\tilde{D}};
$$

$$
\delta_0=\frac{h_0}{\tilde{\delta}};\quad \beta_1=\frac{\delta_0\chi_1\tilde{D}_1}{D_1};\quad \mathrm{Fo}_1=\frac{D_1 l}{u_0 h_0^2}.
$$

In [1, 2] it has been shown that $\delta_0 \to 0$ in cases of nonlinear mass transport of highly soluble gases, i.e., it is possible to leave out of consideration the terms in Eqs. (5) and (6) which are proportional to δ_0.

Mass Transport in the Gas Phase. The rate of mass transfer is limited by mass transport in the gas phase when $\beta_1 \ll 1$. In this case it follows directly from Eq. (6) that

$$
\beta_1=0,\ \left(\frac{\partial C_1}{\partial Y}\right)_{Y=H}=0,\ C_1\equiv 0,
\tag{7}
$$

and the problem reduces to finding a solution of Eq. (5). For this purpose it is necessary to introduce the self-similarity variables

$$
\begin{gathered}
\tilde{U}=\frac{1}{2}\varepsilon\Phi',\ \tilde{V}=\frac{\varepsilon}{2\sqrt{X}}(\eta\Phi'-\Phi),\ \tilde{C}_1=1-\tilde{\Psi},\\
\Phi=\Phi(\eta),\ \tilde{\Psi}=\tilde{\Psi}(\eta),\ \eta=\frac{\tilde{Y}}{2\sqrt{X}}.
\end{gathered}
\tag{8}
$$

Thus, Eq. (5) assumes the form

$$
\tilde{\Psi}''+a\Phi\tilde{\Psi}'=0;\ \ \tilde{\Psi}(0)=1,\ \ \tilde{\Psi}(\infty)=0,
\tag{9}
$$

where $a = \varepsilon_1^2/\varepsilon$, and $\Phi(\eta)$ takes into account the nonlinear mass transport in the gas and is determined in [2] by asymptotic methods. Since Φ depends on two small parameters θ_1 and θ_3, the solution of (9) can be sought in the form of the expansion $\underline{A} = A_0 + \theta_1 A_1 + \theta_3 A_3 + \theta_1^2 A_{11} + \theta_3^2 A_{33} + \theta_1\theta_3 A_{13} + \ldots$, where A is a vector function $A = (\Phi, \tilde{\Psi})$. By using the approximation obtained for Φ in [2] a series of boundary value problems are obtained for $\tilde{\Psi}$.

In the zeroth approximation it is possible to write

$$
\begin{gathered}
\tilde{\Psi}_0''+af(z)\tilde{\Psi}_0'=0;\\
\tilde{\Psi}_0(0)=1,\ \tilde{\Psi}_0(\infty)=0\ \left(z=\frac{2}{\varepsilon}\eta\right).
\end{gathered}
\tag{10}
$$

The solution of Eq. (10) has the form

$$\tilde{\Psi}_0(\eta) = 1 - \frac{1}{\varphi_{01}} \int_0^z E(\varepsilon_1,\ p)dp, \tag{11}$$

where

$$\varphi_{01} = \int_0^\infty E(\varepsilon_1,\ p)\,dp \approx 3{,}01\,\tilde{S}c_1^{-0,35},\quad E(\varepsilon_1,\ p) = \exp\left[-\frac{\varepsilon_1^2}{2}\int_0^p f(s)\,ds\right].$$

To the first approximation with respect to θ_1 it is found that

$$\tilde{\Psi}_1'' + af(z)\tilde{\Psi}_1' = \frac{3a}{\alpha\varepsilon\varphi_{01}} f'(z)E(\varepsilon_1,\ z);$$

$$\tilde{\Psi}_1(0) = \tilde{\Psi}_1(\infty) = 0,$$

i.e.,

$$\tilde{\Psi}_1(\eta) = \frac{3}{2\alpha\varphi_{01}} - \frac{3}{2\alpha\varphi_{01}^2}\int_0^z E(\varepsilon_1,\ p)\,dp - \frac{3}{2\alpha\varphi_{01}} E(\varepsilon_1,\ z), \tag{12}$$

while to the first approximation with respect to θ_3,

$$\tilde{\Psi}_3'' + af(z)\tilde{\Psi}_3' = \frac{4a}{\varepsilon^3\varphi_0\varphi_{01}}\varphi(z)E(\varepsilon_1,\ z);$$

$$\tilde{\Psi}_3(0) = \tilde{\Psi}_3(\infty) = 0,$$

i.e.,

$$\Psi_3(\eta) = \frac{a}{\varepsilon\varphi_0\varphi_{01}}\int_0^z \left[\int_0^p \varphi(s)\,ds\right] E(\varepsilon_1,\ p)\,dp - \frac{a\varphi_{31}}{\varepsilon\varphi_0\varphi_{01}^2}\int_0^z E(\varepsilon_1,\ p)\,dp, \tag{13}$$

where

$$\varphi_{31} = \int_0^\infty \left[\int_0^p \varphi(s)\,ds\right] E(\varepsilon_1,\ p)\,dp \approx 6{,}56\,\tilde{S}c_1^{-0,8}.$$

To the second approximation with respect to θ_1 it is found that

$$\mathbf{\Psi}_{11}'' + af(z)\,\tilde{\Psi}_{11}' = \left[\frac{9a}{2\varepsilon\varphi_{01}}F(z) + \frac{9a}{2\alpha^2\varepsilon\varphi_{01}^2}f'(z) - \right.$$
$$\left. - \frac{9a^2}{4\alpha^2\varphi_{01}} f(z)f'(z)\right]E(\varepsilon_1,\ z);$$

$$\tilde{\Psi}_{11}(0) = \tilde{\Psi}_{11}(\infty) = 0,$$

i.e.

$$\begin{aligned}\tilde{\Psi}_{11}(\eta) = &\left(-\frac{9a\varepsilon\varphi_{21}}{8\varphi_{01}^2} - \frac{9}{4\alpha^2\varphi_{01}^3} + \frac{9a^2\varepsilon^2\varphi_{11}}{32\alpha^2\varphi_{01}^2}\right)\int_0^z E(\varepsilon_1,\ p)\,dp + \\ &+ \frac{9a\varepsilon}{8\varphi_{01}}\int_0^z \left[\int_0^p F(s)\,dz\right]E(\varepsilon_1,\ p)\,dp + \\ &+ \frac{9}{4\alpha^2\varphi_{01}^2}[1 - E(\varepsilon_{11}\,z)] - \frac{9a^2\varepsilon^2}{32\alpha^2\varphi_{01}}\int_0^z f^2(p)E(\varepsilon_1,\ p)\,dp,\end{aligned} \tag{14}$$

where

$$\varphi_{11} = \int_0^\infty f^2(p)E(\varepsilon_1,\ p)\,dp \approx 3{,}011\,\tilde{S}c_1^{-1,608};$$

$$\varphi_{21} = \int_0^\infty \left[\int_0^p F(s)\,ds\right]E(\varepsilon_1,\ p)\,dp \approx 3{,}052\,\tilde{S}c_1^{-1,283}.$$

To the second approximation with respect to θ_3 it is found that

$$\tilde{\Psi}''_{33}+af(z)\,\tilde{\Psi}'_{33}=\left[\frac{4a}{\varepsilon^3\varphi_0^2\varphi_{01}}\left(\frac{\varphi_3}{\varphi_0}+\frac{a\varphi_{31}}{\varepsilon\varphi_{01}}\right)\varphi(z)-\right.$$

$$\left.-\frac{8a}{\varepsilon^5\varphi_0^2\varphi_{01}}\,\bar{\varphi}(z)-\frac{4a^2}{\varepsilon^4\varphi_0^2\varphi_{01}}\,\varphi(z)\int\limits_0^z\varphi(s)\,ds\right]E(\varepsilon_1,\ z);$$

$$\tilde{\Psi}_{33}(0)=\tilde{\Psi}_{33}(\infty)=0,$$

i.e.,

$$\tilde{\Psi}_{33}(\eta)=\left(-\frac{a\varphi_3\varphi_{31}}{\varepsilon\varphi_0^3\varphi_{01}^2}-\frac{a^2\varphi_{31}^2}{\varepsilon^2\varphi_0^2\varphi_{01}^3}+\frac{a^2\varphi_{331}}{2\varepsilon^2\varphi_0^2\varphi_{01}^2}+\right.$$

$$\left.+\frac{2a\bar{\varphi}_{331}}{\varepsilon^3\varphi_0^2\varphi_{01}^2}\right)\int\limits_0^z E(\varepsilon_1,\ p)\,dp+\left(\frac{a\varphi_3}{\varepsilon\varphi_0^3\varphi_{01}}+\right.$$

$$\left.+\frac{a^2\varphi_{31}}{\varepsilon^2\varphi_0^2\varphi_{01}^2}\right)\int\limits_0^z\left[\int\limits_0^p\varphi(s)\,ds\right]E(\varepsilon_1,\ p)\,dp- \tag{15}$$

$$-\frac{a^2}{2\varepsilon^2\varphi_{02}\varphi_1}\int\limits_0^z\left[\int\limits_0^p\varphi(s)\,ds\right]^2E(\varepsilon_1,\ p)\,dp-\frac{2a}{\varepsilon^3\varphi_0^2\varphi_{01}}\int\limits_0^z\left[\int\limits_0^p\bar{\varphi}(s)\,ds\right]E(\varepsilon_1,\ p)\,dp,$$

where

$$\varphi_{331}=\int\limits_0^\infty\left[\int\limits_0^p\varphi(s)\,ds\right]^2E(\varepsilon_1,\ p)\,dp\approx 24\,\tilde{\mathrm{Sc}}_1^{-1,3};$$

$$\bar{\varphi}_{331}=\int\limits_0^\infty\left[\int\limits_0^p\bar{\varphi}(s)\,ds\right]E(\varepsilon_1,\ p)\,dp\approx 0{,}326\,\tilde{\mathrm{Sc}}_1^{-1,63}.$$

The effect of complex interactions is taken into account by the term proportional to $\theta_1\theta_3$:

$$\tilde{\Psi}''_{13}+af(z)\tilde{\Psi}'_{13}=\left[\frac{6a(\varphi_{01}+\varphi_0)}{\alpha\varepsilon^3\varphi_0^2\varphi_{10}^2}\,\varphi(z)-\frac{6a}{\alpha\varepsilon^3\varphi_0\varphi_{01}}\,\bar{\bar{\varphi}}(z)-\right.$$

$$-\frac{3a^2}{\alpha\varepsilon^2\varphi_0\varphi_{01}}\,f'(z)\int\limits_0^z\varphi(s)\,ds+$$

$$\left.+\frac{3a^2\varphi_{31}}{\alpha\varepsilon^2\varphi_0\varphi_{01}^2}\,f'(z)-\frac{3a^2}{\alpha\varepsilon^2\varphi_0\varphi_{01}}\,f(z)\,\varphi(z)\right]E(\varepsilon_1,\ z);$$

$$\tilde{\Psi}_{13}(0)=\tilde{\Psi}_{13}(\infty)=0,$$

i.e.,

$$\tilde{\Psi}_{13}(\eta)=\left[-\frac{3a\varphi_{31}(\varphi_{01}+\varphi_0)}{2\alpha\varepsilon\varphi_0^2\varphi_{01}^2}+\frac{3a\varphi_{131}}{2\alpha\varepsilon\varphi_0\varphi_{01}^2}+\right.$$

$$\left.+\frac{3a\bar{\varphi}_{131}}{2\alpha\varepsilon\varphi_0\varphi_{01}^2}\right]\int\limits_0^z E(\varepsilon_1,\ p)\,dp+\frac{3a(\varphi_{01}+\varphi_0)}{2\alpha\varepsilon\varphi_0^2\varphi_{01}^2}\int\limits_0^z\left[\int\limits_0^p\varphi(s)\,ds\right]E(\varepsilon_1,\ p)\,dp-$$

$$-\frac{3a}{2\alpha\varepsilon\varphi_0\varphi_{01}}\int\limits_0^z\left[\int\limits_0^p\bar{\bar{\varphi}}(s)\,ds\right]E(\varepsilon_1,\ p)\,dp+ \tag{16}$$

$$+\frac{3a}{2\alpha\varepsilon\varphi_0\varphi_{01}}E(\varepsilon_1,\ z)\int\limits_0^z\varphi(s)\,ds-\frac{3a}{2\alpha\varepsilon\varphi_0\varphi_{01}}\int\limits_0^z\varphi(p)\,E(\varepsilon_1,\ p)\,dp+$$

$$+\frac{3a\varphi_{31}}{2\alpha\varepsilon\varphi_0\varphi_{01}^2}\,[1-E(\varepsilon_1,\ z)],$$

where

$$\varphi_{131}=\int\limits_0^\infty\left[\int\limits_0^p\bar{\bar{\varphi}}(s)\,ds\right]E(\varepsilon_1,\ p)\,dp\approx\tilde{\mathrm{Sc}}_1^{-1,3};$$

$$\bar{\varphi}_{131}=\int_0^\infty \varphi(p)\,E(\varepsilon_1,\,p)\,dp\approx 4{,}18\,\tilde{\mathrm{Sc}}_1^{-0{,}46}.$$

The expressions (11)-(16) make it possible to determine the Sherwood number. In order to do this it is necessary to substitute Eqs. (2) and (4) into Eq. (3). In this way, it is found that

$$\tilde{\mathrm{Sh}}_1=\sqrt{\tilde{\mathrm{Pe}}}\left[-\int_0^1\left(\frac{\partial\tilde{C}_1}{\partial\tilde{Y}}\right)_{\tilde{Y}=0}dX-\theta_3\frac{\tilde{D}}{\tilde{D}_1}\frac{\chi_1c_{10}}{\tilde{c}_{10}-\chi_1c_{10}}\int_0^1\left(\frac{\partial\tilde{C}}{\partial\tilde{Y}}\right)_{\tilde{Y}=0}dX\right].$$

In the case of a highly soluble gas $\chi_1 \to 0$, i.e., the second term in the expression for the Sherwood number is unimportant, so that

$$\tilde{\mathrm{Sh}}_1=-\sqrt{\tilde{\mathrm{Pe}}}\int_0^1\left(\frac{\partial\tilde{C}_1}{\partial\tilde{Y}}\right)_{\tilde{Y}=0}dX=\sqrt{\tilde{\mathrm{Pe}}}\,\tilde{\Psi}'(0), \tag{17}$$

where $\tilde{\mathrm{Pe}} = \tilde{u}_0\ell/\tilde{D}$, and $\Psi'(0)$ is defined by the expression

$$\begin{aligned}\tilde{\Psi}'(0)=&-\frac{2}{\varepsilon\varphi_{01}}-\theta_1\frac{3}{\alpha\varepsilon\varphi_{01}^2}-\theta_3\frac{2a\varphi_{13}}{\varepsilon^2\varphi_0\varphi_{01}^2}-\\&-\theta_1^2\left(\frac{9a\varphi_{21}}{4\varphi_{01}^2}+\frac{9}{2\alpha^2\varepsilon\varphi_{01}^3}-\frac{9a^2\varepsilon\varphi_{11}}{16\alpha^2\varphi_{01}^2}\right)-\\&-\theta_3^2\left(\frac{2a\varphi_3\varphi_{31}}{\varepsilon^2\varphi_0^3\varphi_{01}^2}+\frac{2a^2\varphi_{31}^2}{\varepsilon^3\varphi_0^2\varphi_{01}^3}-\frac{a^2\varphi_{331}}{\varepsilon^3\varphi_0^2\varphi_{01}^2}-\frac{4a\bar{\varphi}_{331}}{\varepsilon^4\varphi_0^2\varphi_{01}^2}\right)-\\&-\theta_1\theta_3\frac{3a}{\alpha\varepsilon^2\varphi_0\varphi_{01}^2}\left[\varphi_{31}\left(1+\frac{\varphi_{01}}{\varphi_0}\right)+\varphi_{131}+\bar{\varphi}_{131}\right].\end{aligned} \tag{18}$$

Comparison of $\tilde{\Psi}'(0)$ with $\Psi'(0)$ (obtained in [2]) shows that it differs only slightly because of the small differences in the values of ε_1 and ε. It follows from this that in cases of multicomponent mass transport in which the mass transport of one component is nonlinear as a result of a large concentration gradient, the mass transport coefficients of all the other components will have similar values.

Mass Transport in the Liquid Phase. The rate of mass transfer is limited by the mass transport in the liquid phase when $\beta_1 \gg 1$. In this cae it follows directly from Eq. (6) that

$$\beta_1^{-1}=0,\quad\left(\frac{\partial\tilde{C}_1}{\partial\tilde{Y}}\right)_{\tilde{Y}=0}=0,\quad\tilde{C}_1\equiv 1,\quad(C_1)_{Y=H}=(\tilde{C}_1)_{\tilde{Y}=0}=1, \tag{19}$$

and the problem reduces to solving Eq. (6) with the conditions given by Eq. (19) and U, V, and H as defined in [2].

In order to solve Eq. (6) it is necessary to introduce the diffusion boundary layer variable

$$Y_1=\frac{H-Y}{\sqrt{\mathrm{Fo}_1}}. \tag{20}$$

Equation (6) then assumes the form

$$\begin{gathered}U\frac{\partial C_1}{\partial X}+\frac{H'U-V}{\sqrt{\mathrm{Fo}_1}}\frac{\partial C_1}{dY_1}=\frac{\partial^2C_1}{\partial Y_1^2};\\X=0,\quad C_1=0;\\Y_1=0,\quad C_1=1;\\Y_1\to\infty,\quad C_1=0.\end{gathered} \tag{21}$$

If the following notation is used,

$$\delta_1 = \mathrm{Fo}_1,\ \delta_2 = \theta_2,\ \delta_3 = \theta_0\theta_3,\ \delta_4 = \frac{\theta_0\theta_3}{\sqrt{\mathrm{Fo}_1}}, \tag{22}$$

then to a first approximation with respect to the small parameters δ_i $(i = 1,\dots,4)$ the problem (21) can be written as

$$\left[\frac{3}{2}(1-\delta_1 Y_1^2)+\delta_2\frac{\alpha}{2\varepsilon\sqrt{X}}-\delta_3\frac{2}{\varepsilon\varphi_0}(\sqrt{L_\infty}-\sqrt{X})\right]\frac{\partial C_1}{\partial X}+$$
$$+\left[\delta_2\frac{\alpha Y_1}{4\varepsilon X\sqrt{X}}-\delta_3\frac{Y_1}{\varepsilon\varphi_0\sqrt{X}}+\delta_4\frac{1}{\varepsilon\varphi_0\sqrt{X}}\right]\frac{\partial C_1}{\partial Y_1}=\frac{\partial^2 C_1}{\partial Y_1^2}; \tag{23}$$
$$X = 0,\quad C_1 = 0;$$
$$Y_1 = 0,\quad C_1 = 0;$$
$$Y_1 \to \infty,\quad C_1 = 0.$$

For solving the problem (23) it is necessary to use the expansion

$$C_1 = C_{10} + \delta_1 C_{11} + \delta_2 C_{12} + \delta_3 C_{13} + \delta_4 C_{14} + \cdots . \tag{24}$$

Thus, by using the perturbation method a series of boundary value problems is obtained. To the zeroth approximation it is possible to write:

$$\frac{3}{2}\frac{\partial C_{10}}{\partial X}=\frac{\partial^2 C_{10}}{\partial Y_1^2};$$
$$X = 0,\quad C_{10} = 0;$$
$$Y_1 = 0,\quad C_{10} = 1;$$
$$Y_1 \to \infty,\quad C_{10} = 0,$$

i.e.,

$$C_{10} = \operatorname{erfc}\zeta,\quad \zeta = \left(\frac{3Y_1^2}{8X}\right)^{1/2}. \tag{25}$$

To the first approximation with respect to δ_1 it is found that

$$\frac{3}{2}\frac{\partial C_{11}}{\partial X}=\frac{\partial^2 C_{11}}{\partial Y_1^2}+\frac{3}{2}Y_1^2\frac{\partial C_{10}}{\partial X};$$
$$X = 0,\quad C_{11} = 0;$$
$$Y_1 = 0,\quad C_{11} = 0;$$
$$Y_1 \to \infty,\quad C_{11} = 0,$$

where C_{11} is determined [6] by the Green's function method:

$$C_{11}=\frac{1}{\sqrt{6\pi}}\left(Y_1\sqrt{X}+\frac{Y_1^3}{2\sqrt{X}}\right)\exp\left(-\frac{3Y_1^2}{8X}\right). \tag{26}$$

To the first approximation with respect to δ_2 it is possible to write

$$\frac{3}{2}\frac{\partial C_{12}}{\partial X}-\frac{\partial^2 C_{12}}{\partial Y_1^2}=\frac{\alpha}{2\varepsilon\sqrt{X}}\left(\frac{\partial C_{10}}{\partial X}+\frac{Y_1}{2\sqrt{X}}\frac{\partial C_{10}}{\partial Y_1}\right);$$
$$X = 0,\quad C_{12} = 0;$$
$$Y_1 = 0,\quad C_{12} = 0;$$
$$Y_1 \to \infty,\quad C_{12} = 0,$$

where the right-hand part is equal to zero, i.e.,

$$C_{12} \equiv 0. \tag{27}$$

To the first approximation with respect to δ_3 it is found that

$$\frac{3}{2}\frac{\partial C_{13}}{\partial X}=\frac{\partial^2 C_{13}}{\partial Y_1^2}+\frac{2}{\varepsilon\varphi_0}(\sqrt{L_\infty}-\sqrt{X})\frac{\partial C_{10}}{\partial X}+\frac{Y_1}{\varepsilon\varphi_0\sqrt{X}}\frac{\partial C_{10}}{\partial Y_1};$$

$$X = 0, \quad C_{13} = 0;$$
$$Y_1 = 0, \quad C_{13} = 0;$$
$$Y_1 \to \infty, \quad C_{13} = 0,$$

where C_{13} can be found by the Green's function method:

$$C_{13} = \sqrt{\frac{2}{3\pi}} \frac{1}{\varepsilon\varphi_0} \left(\sqrt{L_\infty} \frac{Y_1}{\sqrt{X}} - \frac{4}{3} Y_1 \right) \exp\left(-\frac{3Y_1^2}{8X} \right). \tag{28}$$

To the first approximation with respect to δ_4 it is possible to write

$$\frac{3}{2} \frac{\partial C_{14}}{\partial X} = \frac{\partial^2 C_{14}}{\partial Y_1^2} - \frac{1}{\varepsilon\varphi_0 \sqrt{X}} \frac{\partial C_{10}}{\partial Y_1};$$
$$X = 0, \quad C_{14} = 0;$$
$$Y_1 = 0, \quad C_{14} = 0;$$
$$Y_1 \to \infty, \quad C_{14} = 0,$$

where C_{14} is obtained by using Green's function:

$$C_{14} = \frac{2}{\varepsilon\varphi_0} \sqrt{\frac{2}{3\pi}} \left[\exp\left(-\frac{3Y_1^2}{8X} \right) - \operatorname{erfc}\left(\frac{3Y_1^2}{8X} \right)^{1/2} \right]. \tag{29}$$

Equations (25)-(29) make it possible to determine the Sherwood number. For this purpose it is possible to find directly from Eqs. (2)-(4), (20), and [1] that

$$\begin{gathered} \mathrm{Sh}_1 = \sqrt{\mathrm{Pe}_1} \int_0^1 \left(\frac{\partial C_1}{\partial Y_1} \right)_{Y_1=0} dX - \\ - \frac{\tilde{c}_0 - \chi c_0}{\chi} \frac{MDl\beta}{\rho_0^* D_1 h_0} \left[\frac{\chi_1 c_{10}}{\tilde{c}_{10} - \chi_1 c_{10}} \int_0^1 \left(\frac{\partial \tilde{C}}{\partial \tilde{Y}} \right)_{\tilde{Y}=0} dX + \right. \\ \left. + \int_0^1 (C_1)_{Y_1=0} \left(\frac{\partial \tilde{C}}{\partial \tilde{Y}} \right)_{\tilde{Y}=0} dX \right] \approx \sqrt{\mathrm{Pe}_1} \int_0^1 \left(\frac{\partial C_1}{\partial Y_1} \right)_{Y_1=0} dX, \end{gathered} \tag{30}$$

where $\mathrm{Pe}_1 = u_0 \ell / D_1$. From Eqs. (25)-(30) it is found finally that

$$\begin{gathered} \mathrm{Sh}_1 = -\sqrt{\frac{6\mathrm{Pe}_1}{\pi}} \left[1 - \frac{\delta_1}{9} - \frac{2\delta_3}{3\varepsilon\varphi_0} \left(\sqrt{L_\infty} - \frac{2}{3} \right) - \right. \\ \left. - \frac{2\delta_4}{\varepsilon\varphi_0} \sqrt{\frac{2}{3\pi}} \right]. \end{gathered} \tag{31}$$

It can be seen from Eq. (31) that the parameters δ_3 and δ_4 depend on the concentration gradient of the component with intensive mass transfer:

$$\delta_3 = \frac{\tilde{\rho}^*}{\tilde{\rho}_0^*} \frac{\sqrt{\tilde{D} \tilde{u}_0 l}}{u_0 h_0} \frac{M (\tilde{c}_0 - \chi c_0)}{\rho_0^*},$$

$$\delta_4 = \frac{\tilde{\rho}^*}{\tilde{\rho}_0^*} \sqrt{\frac{\tilde{D} \tilde{u}_0}{D_1 u_0}} \frac{M (\tilde{c}_0 - \chi c_0)}{\rho_0^*},$$

i.e., that the rate of mass transport of a component in the liquid is determined by the concentration gradient of the component with intensive mass transfer and depends on the direction of this mass transfer.

The absorption (desorption) of the component with intensive mass transfer leads to a decrease (increase) in the rates of mass transport of the remaining components in the liquid. By comparing Eqs. (18) and (31) it can be seen that the effect of the direction of the intensive mass transfer is opposite in cases when the mass transfer of the components with the small concentration gradients is limited by mass transport in the gas or in the liquid.

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EFFECT OF THE CONCENTRATION DEPENDENCE OF THE DIFFUSION COEFFICIENT ON THE DISTRIBUTION AND FLOW OF HYDROGEN IN PALLADIUM MEMBRANES

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UDC 539.219.3:669.234.788

Results are presented from the calculation of steady-state profiles of the distribution and flow of hydrogen in palladium membranes. The distributions are obtained with allowance for features of the dependence of the diffusion coefficient on hydrogen concentration.

The study [1] examined the effect of a nonlinear hydrogen distribution through the thickness of metallic membranes, due to the concentration dependence of the diffusion coefficient, on the permeability of the membranes to hydrogen. The study made use of a simple model relation D(c) [2] which describes phase transformation in hydride-forming metals [3]. However, it has become evident in recent years that along with a minimum corresponding to phase transformation, the relation D(c) for Pd–H systems has a maximum in the concentration range c = (0.6...0.7)H/Pd [4-7]. In the present study, we discuss the results of calculation of steady-state profiles of hydrogen distribution in palladium membranes and the flow of hydrogen through these membranes with allowance for a relation D(c) that reflects the above features.

In accordance with [7], the concentration dependence of the coefficient of diffusion of H in Pd is approximately described by the relation

$$D(c) = D(0)\left[1-6\frac{\beta-1}{\beta}+\left(\frac{\partial\mu_e}{\partial c}-\frac{3\gamma}{2}\,\mathrm{cth}\,\frac{\hbar\omega_\alpha-\gamma c}{2kT}\right)\frac{c(1-c)}{kT}\right], \quad (1)$$

where

$$\beta=\sqrt{1+4\alpha c(1-c)}, \quad \alpha=\exp(-w/kT)-1, \quad (2)$$

while the electronic contribution to the chemical potential of the hydrogen subsystem $\mu_e(c)$ is determined from the empirical formula [3, Vol. 2, p. 152]:

$$\mu_e(c)=\sum_{n=1}^{5}(-1)^{n-1}a_n c^n, \quad c<0{,}656, \quad (3)$$

$$\mu_e(c)=b(c-c_1), \qquad c\geqslant 0{,}656.$$

Donetsk Polytechnic Institute. Translated from Inzhenerno-fizicheskii Zhurnal, Vol. 59, No. 10, pp. 602-606, October, 1990. Original article submitted September 5, 1989.