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

1. NUMERICAL ANALYSIS

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A numerical analysis is carried out of the kinetics of nonlinear mass transfer between a gas and a falling liquid film. The nonlinear effect which is considered is the result of intensive mass transfer. Under these conditions the large mass flux induces a secondary flow at the phase interface. The mass transport in both phases takes into account the influence of the intensive mass transfer on the hydrodynamic behavior. An equation is obtained for the rate of absorption is greater than the rate of desorption under conditions of intensive mass transfer.

Theoretical investigations of mass transport in two-phase systems with intensive mass transfer between the phases have shown [1, 2] that large mass fluxes through the phase interface induce secondary flows. The velocity of such secondary flows (normal to the interfacial surface) is the reason for the appearance of nonlinear effects in the kinetics of mass transport.

The analysis of nonlinear mass transport in falling liquid films [3, 4] (with chemical reactions in the liquid) has shown that the nonlinear effects occur only at large pressures. At small pressures, the effect of the secondary flow cannot be kept track of, and there remain only the nonlinear effects arising because of the concentration dependence of the diffusion coefficients. This is explained by the fact that the velocity of the secondary flow increases due to the intensification of the mass transfer as a result of the increase in the rate of the chemical reaction in the liquid. On the other hand, the increase in the rate of the overall reaction leads to a decrease in the contribution of the convective transport to the overall rate of mass transport. Thus, the chemical reaction in the liquid phase intensifies mass transfer and causes the secondary flow, but this flow (like the basic flow) does not have an influence on mass transport in the liquid.

Theoretical investigations of nonlinear mass transport in gas-liquid systems have shown [7, 8] that in practice the nonlinear effects are significant when the mass transfer process is limited by the mass transport in the gas phase. In the case where the diffusional resistances are comparable in both phases, the nonlinear effects are appreciably reduced, and when the process is limited by the diffusional resistance in the liquid they practically disappear (at normal pressures).

A review of work on the kinetics of nonlinear mass transport in two-phase systems with intensive mass transfer indicates the considerable importance of the problem of setting up a theory of nonlinear mass transfer between gases and falling liquid films in cases where the process is limited by mass transport in the gas phase, and this is the objective of the present paper. On this basis attempts are made to explain the differences in the rates of mass transport which are observed experimentally during the absorption and desorption of very soluble gases.

<u>Mathematical Description</u>. Nonlinear mass transfer between a gas and a falling liquid film during the absorption and desorption of the gas can be described using the equations of motion and the convective diffusion in the liquid and the gas. The form of the surface of the film is obtained from the equation for the macroscopic balance for the liquid in the film, i.e., from the condition for the "nonpassage" of the liquid through the surface of the film. In the case of intensive mass transfer it is necessary to introduce the velocity

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of the secondary flow as a function of the mass flux. The velocity of the secondary flow is a velocity component normal to the interfacial surface. Thus, the dependence of the velocity of the secondary flow on the mass flux expresses the condition of "passage" through the interfacial boundary as a result of the intensive mass transfer.

Theoretical and experimental investigations [1, 2] show that the flow of a liquid film on a vertical surface can be described with sufficient accuracy by the equations of motion to the zeroth approximation with respect to the small parameter $(h_0/\ell)^2$ (which will be observed below and in all the remaining equations):

$$v \frac{\partial^2 u}{\partial y^2} + g = 0, \quad \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0.$$
 (1)

The boundary conditions for the system (1) express the "adherence" of the liquid on the solid surface at y = 0 and the continuity of the stress tensor at the phase interface at y = h:

$$y = 0, \quad u = 0, \quad v = 0,$$

 $y = h, \quad \mu \frac{\partial u}{\partial y} = \tilde{\mu} \frac{\partial \tilde{u}}{\partial y}.$
(2)

The film thickness is determined [1, 2] from the condition for "passage" through the phase boundary, i.e., from the macroscopic balance for the liquid in the film taking into account the secondary flow [3] as a result of the intensive mass transfer from the gas:

$$y = h, \quad h' u - v = \frac{MD}{\rho_0^*} \frac{\partial c}{\partial y}. \tag{3}$$

Changes in the film thickness are the result of changes in the liquid velocity over the length of the film. This leads to the need to introduce into the boundary condition for the velocity the thickness over the length of the film. A theoretical analysis of film flow has shown [1, 2, 9] that all the effects become rapidly attenuated because of the small thickness of the film, and that at some distance ℓ_{∞} from the inlet of the film asymptotic flow is found, i.e., the velocity of the flow no longer depends on the longitudinal coordinate. This provides the basis for using

 $x \to l_{\infty}, \quad \frac{\partial u}{\partial x} \to 0, \quad h \to h_0$ (4)

as a boundary condition for Eqs. (1) and (3).

In order to determine the concentration of the material being absorbed (desorbed) in Eq. (3) it is necessary to solve the equation of convective diffusion in the liquid. In practice the thickness of the diffusional boundary layer in the liquid is always smaller than the thickness of the film, so that it is possible to use the boundary layer approximation:

$$u \frac{\partial c}{\partial x} + v \frac{\partial c}{\partial y} = D \frac{\partial^2 c}{\partial y^2}$$
(5)

with boundary conditions which take into account the impermeability of the solid wall and the continuity of the mass flux through the phase boundary, where the mass flux has a convective component as a result of the secondary flow:

$$x = 0 c = c_0;$$

$$y = 0 \frac{\partial c}{\partial y} = 0;$$
(6)

$$y = h$$
 $\frac{D\rho^*}{\rho_0^*} \frac{\partial c}{\partial y} = \frac{D\rho^*}{\tilde{\rho}_0^*} \frac{\partial c}{\partial y},$

and where at ordinary pressures $\rho^*/\rho \stackrel{*}{_0} \approx 1$.

Theoretical analyses of the simultaneous flow of the gas and the liquid film [1, 2, 10] show that the flow of the gas can be described using the boundary layer approximation:

$$\tilde{u} \ \frac{\partial \tilde{u}}{\partial x} + \tilde{v} \ \frac{\partial \tilde{u}}{\partial y} = \tilde{v} \ \frac{\partial^2 \tilde{u}}{\partial y^2} \ , \ \frac{\partial \tilde{u}}{\partial x} + \frac{\partial \tilde{v}}{\partial y} = 0.$$
(7)

The boundary conditions for Eq. (7) express the continuity of the velocity at the phase interface and the potential motion of the gas beyond the boundary layer:

$$x = 0 \qquad \tilde{u} = \tilde{u}_{0};$$

$$y = h \qquad \tilde{u} = u, \qquad \tilde{v} = v;$$

$$y \to \infty \qquad \tilde{u} = \tilde{u}_{0}.$$
(8)

In gases the thicknesses of the laminar and diffusional boundary layers have the same order of magnitude, and for describing the mass transport in the gas phase it is possible to use an analogous approximation

$$\tilde{u} \quad \frac{\partial \tilde{c}}{\partial x} + \tilde{v} \quad \frac{\partial \tilde{c}}{\partial u} = \tilde{D} \frac{\partial^2 \tilde{c}}{\partial u^2} \tag{9}$$

with boundary conditions expressing the thermodynamic equilibrium at the phase interface and the constancy of the concentration beyond the boundary layer:

$$\begin{aligned} x &= 0 & \tilde{c} = \tilde{c}_0; \\ y &= h & \tilde{c} = \chi c; \\ y &\to \infty & \tilde{c} = \tilde{c}_0. \end{aligned}$$
 (10)

Intensive mass transfer between the gas and liquid induces secondary flow [1, 2, 7], and its velocity \tilde{v}_n (to the approximation indicated above) can be expressed in terms of the mass flux through the phase interface:

$$\tilde{v}_n = \tilde{v}(x, h) - h'\tilde{u}(x, h) = -\frac{M\tilde{D}}{\tilde{\rho}_0^*} \left(\frac{\partial\tilde{c}}{\partial y}\right)_{y=h}.$$
(11)

Equation (11) expresses the effect of mass transport on the hydrodynamics, and provides the reason for the appearance of nonlinearity in the left side of Eq. (9), i.e., Eq. (11) is the basis for calculating the nonlinear mass transport under conditions of intensive mass transfer.

<u>The Kinetics of Mass Transport.</u> In the case being considered the rate of mass transfer between the gas and the liquid is determined by the kinetics of the nonlinear mass transport in the gas phase. This rate can be determined by averaging the mass flux over the length ℓ of the film:

$$J = M\tilde{k}(\tilde{c}_0 - \chi c_0) = \frac{1}{l} \int_0^l \tilde{I} \, dx,$$
 (12)



Fig. 1. The dimensionless velocity (Φ '), concentration (Ψ), and diffusion flux (Ψ ') in the diffusion boundary layer in the gas ($\varepsilon = 1$) for $\theta_1 = 0.07$ and various values of θ : 1): $\theta = -0.3$; 2): $\theta = -0.2$; 3) $\theta = -0.1$; 4) $\theta = 0$; 5): $\theta = 0.1$; 6) $\theta = 0.2$; 7) $\theta = 0.3$.

where the local mass flux has a diffusional and a convective component

$$\tilde{I} = -M\tilde{D}\left(\frac{\partial \tilde{c}}{\partial y}\right)_{y=h} + M\chi c_0 \left[\tilde{v}(x, h) - h' u(x, h)\right] = -\frac{M\tilde{D}\tilde{\rho}^*}{\tilde{\rho}^*_0} \left(\frac{\partial \tilde{c}}{\partial y}\right)_{y=h}.$$
(13)

The expressions (12) and (13) make it possible to determine the Sherwood number after solving Eqs. (1)-(11):

$$\tilde{Sh} = \frac{\tilde{k}l}{\tilde{D}} = -\frac{\tilde{\rho}^*}{\tilde{\rho}_0^*} \frac{1}{\tilde{c}_0 - \chi c_0} \int_0^l \left(\frac{\tilde{\partial c}}{\partial y}\right)_{y=h} dx.$$
 (14)

<u>Method of Solution</u>. In order to solve the system of equations (1)-(11) it is necessary to introduce the following dimensionless variables:

$$X = \frac{x}{l}, \quad Y = \frac{y}{h_0}, \quad \tilde{Y} = \frac{y-h}{\tilde{\delta}}, \quad H(X) = \frac{h}{h_0},$$

$$U(X, Y) = \frac{u}{u_0}, \quad V(X, Y) = \frac{v}{\varepsilon_0 u_0}, \quad C(X, Y) = \frac{c-c_0}{\tilde{c_0}/\chi - c_0},$$

$$\tilde{U}(X, \tilde{Y}) = \frac{\tilde{u}}{\tilde{u}_0}, \quad \tilde{V}(X, \tilde{Y}) = \frac{\tilde{v}}{\tilde{\varepsilon_0}\tilde{u}_0}, \quad \tilde{C}(X, \tilde{Y}) = \frac{\tilde{c}-\chi c_0}{\tilde{c_0}-\chi c_0},$$
(15)

where

$$\varepsilon_0 = \frac{h_0}{l}; \quad u_0 = \frac{gh_0^2}{3v}; \quad \tilde{\varepsilon}_0 = \frac{\tilde{\delta}}{l}; \quad \tilde{\delta} = \sqrt{\frac{\tilde{D}l}{\tilde{u}_0}}. \quad (16)$$

In this way it is found that

$$\frac{\partial^2 U}{\partial Y^2} = -3, \quad \frac{\partial U}{\partial X} + \frac{\partial V}{\partial Y} = 0;$$

$$Y = 0, \quad U = 0, \quad V = 0;$$

$$Y = H, \quad \frac{\partial U}{\partial Y} = \theta_2 \left(\frac{\partial \tilde{U}}{\partial \tilde{Y}}\right)_{\tilde{Y}=0};$$

$$X \to L_{\infty}, \quad \frac{\partial U}{\partial X} \to 0;$$

$$H' U (X, H) = V (X, H) + \theta_4 \left(\frac{\partial C}{\partial Y}\right)_{Y=H};$$

$$X \to L_{\infty}, \quad H \to 1;$$
(18)

$$U = \frac{\partial C}{\partial X} + V \frac{\partial C}{\partial Y} = Fo \quad \frac{\partial^2 C}{\partial Y^2};$$

$$X = 0, \quad C = 0;$$

$$Y = 0, \quad \frac{\partial C}{\partial Y} = 0;$$
(19)

$$Y = H, \quad \frac{\partial C}{\partial Y} = \beta \left(\frac{\partial \tilde{C}}{\partial \tilde{Y}} \right)_{\tilde{Y}=0};$$

$$\widetilde{U} \frac{\partial \widetilde{U}}{\partial X} + \widetilde{V} \frac{\partial \widetilde{U}}{\partial \widetilde{Y}} = \widetilde{S}c \ \frac{\partial^2 \widetilde{U}}{\partial \widetilde{Y}^2} + \delta_0 H' \widetilde{U} \ \frac{\partial \widetilde{U}}{\partial \widetilde{Y}},$$

$$\frac{\partial \widetilde{U}}{\partial X} + \frac{\partial \widetilde{V}}{\partial \widetilde{Y}} = \delta_0 H' \ \frac{\partial \widetilde{U}}{\partial \widetilde{Y}};$$

$$X = 0, \ \widetilde{U} = 1;$$

$$\widetilde{Y} = 0, \ \widetilde{U} = \theta_1 U(X, H), \ \widetilde{V} = -\theta_3 \ \frac{\partial \widetilde{C}}{\partial \widetilde{V}} + \delta_0 H' \widetilde{U};$$
(20)

$$\begin{split} \partial Y \\ \tilde{Y} \to \infty, \ \tilde{U} = 1; \\ \tilde{U} \frac{\partial \tilde{C}}{\partial X} + \tilde{V} \frac{\partial \tilde{C}}{\partial \tilde{Y}} &= \frac{\partial^2 \tilde{C}}{\partial \tilde{Y}^2} + \delta_0 H' \tilde{U} \frac{\partial \tilde{C}}{\partial \tilde{Y}}; \\ X = 0, \ \tilde{C} = 1; \\ \tilde{Y} = 0, \ \tilde{C} = 0; \\ \tilde{Y} \to \infty, \ \tilde{C} = 1, \end{split}$$
(21)

$$L_{\infty} = \frac{l_{\infty}}{l}; \quad \delta_0 = \frac{h_0}{\tilde{\delta}}; \quad Fo = \frac{Dl}{u_0 h_0}; \quad \tilde{S}c = \frac{\tilde{v}}{\tilde{D}};$$

$$\theta_1 = \frac{u_0}{\tilde{u}_0}; \quad \theta_2 = \frac{\tilde{\mu}\delta_0}{\mu\theta_1}; \quad \theta_3 = \frac{M}{\tilde{\rho}_0^*} (c_0 - \chi c_0);$$

$$\theta_4 = \frac{MD(\tilde{c}_0 - \chi c_0)}{\varepsilon_0 u_0 h_0 \rho_0^* \chi}; \quad \beta = \frac{\delta_0 \chi \tilde{D} \tilde{\rho}^*}{\tilde{D} \tilde{\rho}_0^*}.$$

(22)

The parameter θ_3 determines the direction of mass transfer [12]. In the case of absorption, $\theta_3 > 0$, while for desorption, $\theta_3 < 0$. To the approximation of the linear theory of mass transport $\theta_3 = 0$ [11].

The solution of the problem (17)-(21) makes it possible to determine the Sherwood number from Eq. (14), which in terms of the new variables has the form

$$\vec{\mathrm{Sh}} = -\frac{\vec{\rho}^*}{\vec{\rho}_0^*} \sqrt{\vec{\mathrm{Pe}}} \int_0^1 \left(\frac{\partial \vec{C}}{\partial \vec{Y}}\right)_{\vec{Y}=0} dX, \ \vec{\mathrm{Pe}} = \frac{\vec{u}_0 l}{\vec{D}}.$$
 (23)

In the case of a highly soluble gas, when the process is limited by mass transport in the gas phase, the parameters β and δ_0 in Eqs. (17)-(21) are practically equal to zero. If $\beta = \delta_0 = 0$ is substituted into Eqs. (17)-(21), it follows directly from Eq. (19) that $C \equiv 0$, and in Eq. (18) it is necessary to substitute

$$\theta_{4} \left(\frac{\partial C}{\partial Y} \right)_{Y=H} = \theta_{0} \theta_{3} \left(\frac{\partial \tilde{C}}{\partial \tilde{Y}} \right)_{\tilde{Y}=0}, \quad \theta_{0} = \frac{\tilde{\rho}^{*} V \tilde{u}_{0} \tilde{D} l}{h_{0} u_{0} \rho_{0}^{*}},$$

using the condition for the continuity of the mass flux through the phase interface. In this approximation the system of equations (17), (18), (20), and (21) can be broken down into two apparently independent problems for the liquid and the gas.

<u>Velocity Distributions and Thicknesses of the Films</u>. The velocity distribution in the film is determined directly from Eq. (17):

$$U = -\frac{3}{2}Y^{2} + \left[3H + \theta_{2}\left(\frac{\partial \tilde{U}}{\partial \tilde{Y}}\right)_{\tilde{Y}=0}\right]Y,$$

$$V = -\frac{1}{2}\left[3H' + \theta_{2}\frac{d}{dX}\left(\frac{\partial \tilde{U}}{\partial \tilde{Y}}\right)_{\tilde{Y}=0}\right]Y^{2}.$$
(24)

The film thickness is found from Eq. (18) after the substitution of Eq. (24):

$$H^{3} = 1 - \frac{1}{2} \theta_{2} \left(\frac{\partial \tilde{U}}{\partial \tilde{Y}} \right)_{\tilde{Y}=0} H^{2} - \theta_{0} \theta_{3} \int_{X}^{L_{\infty}} \left(\frac{\partial \tilde{C}}{\partial \tilde{Y}} \right)_{\tilde{Y}=0} dX.$$
(25)

The final expressions for determining the velocity (Eq. (24)) and the film thickness (Eq. (25)) are obtained after solving Eqs. (20) and (21).

Velocity and Concentration Distributions in the Gas. In Eq. (24) θ_2 is a small parameter, i.e., it is possible to substitute \tilde{U} in the zeroth approximation with respect to θ_2 . This makes it possible to solve Eq. (20) to the zeroth approximation with respect to θ_2 . For this purpose it is necessary to substitute into Eq. (20) U(X, H) to the zeroth approximation with respect to the small parameter θ_2 , i.e.,

$$U(X, H) = \frac{3}{2}.$$
 (26)

This relationship is obtained from Eqs. (24) and (25) if $\theta_2 = 0$ is substituted.

The velocity and concentration distributions in the gas are obtained from Eq. (20) and (21) by means of the self-similarity variables

$$\begin{split} \tilde{U} &= \frac{1}{2} \, \epsilon \Phi', \quad V = \frac{\epsilon}{2} \, \frac{1}{\sqrt{X}} \, (\eta \Phi' - \Phi), \quad \tilde{C} = 1 - \Psi, \\ \Phi &= \Phi \, (\eta), \quad \Psi = \Psi \, (\eta), \quad \eta = \frac{\tilde{Y}}{2 \, \sqrt{X}}, \quad \epsilon = \sqrt{\tilde{S}c} \, . \end{split}$$

Thus, it follows directly from Eqs. (20) and (21) that

$$\Phi^{\prime\prime\prime} + \frac{1}{\varepsilon} \Phi \Phi^{\prime\prime} = 0, \quad \Psi^{\prime\prime} + \varepsilon \Phi \Psi^{\prime} = 0,$$

$$\Phi(0) = -\frac{\theta_3}{\varepsilon} \Psi^{\prime}(0), \quad \Phi^{\prime}(0) = \frac{3\theta_1}{\varepsilon}, \quad \Phi^{\prime}(\infty) = \frac{2}{\varepsilon},$$

$$\Psi(0) = 1, \quad \Psi(\infty) = 0.$$
(27)

The rate of mass transport is determined by the Sherwood number from Eq. (23). In terms of the new variables this expression assumes the form

$$\tilde{Sh} = \frac{\tilde{\rho}^*}{\rho_0^*} \, \sqrt{\tilde{Pe}} \, \Psi' \, (0), \tag{28}$$

where $\overline{\Psi}'(0)$ is found from the solution of the problem (27). This solution was found by numerical means for $\varepsilon = 1$ and various values of the parameters θ_1 and θ_3 (see Fig. 1).

<u>Conclusions</u>. The results which have been obtained show that the rate of mass transport depends significantly on the direction of mass transfer. In cases of the absorption of highly soluble gases with large concentration gradients the rate of mass transport exceeds the rate of desorption. This effect cannot be predicted from the linear theory of mass transport.

The kinetics of nonlinear mass transfer depend significantly on the Schmidt number in the gas phase (ε), i.e., it is necessary to solve the problem (27) for each specific case. Finding this solution is made difficult as the result of the presence of boundary conditions at infinity. It is therefore expedient to find an asymptotic solution of the problem (27) using the condition that the parameters θ_1 and θ_3 are small for cases of practical interest.

NOTATION

c, concentration; c*, concentration at phase interface; D, diffusion coefficient; 3, acceleration of free fall; h, film thickness; h_0 , film thickness according to Nusselt theory; I, local mass flux; J, rate of mass transfer; k, mass transfer coefficient; ℓ , length of film; M, molecular mass; u, longitudinal component of velocity; v, transverse component of velocity; x, longitudinal coordinate; y, transverse coordinate; v, kinematic coefficient of viscosity; ρ_0 , density of pure liquid; $\rho^* = \rho_0^* + Mc^*$, density of solution of liquid at phase interface; χ , Henry's constant.

SUBSCRIPTS AND SUPERSCRIPTS

~, denotes quantity in gas; 0, characteristic values of quantities in liquid and gas;
', ", "', derivative functions.

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TWO-PHASE MULTICOMPONENT MASS TRANSPORT IN A DESCENDING, STRAIGHT-THROUGH FLOW OF PHASES

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The parameters of combined, multicomponent mass transport in a two-phase gas (vapor)-liquid system moving as a descending, laminar straight-through flow are investigated theoretically, and methods for calculating them are proposed. The methods are based on solving the system of differential equations for multi-component convective diffusion in both phases with an allowance for the conjugation conditions at interface. The diffusion equations are solved both numerically and analytically in the boundary-layer approximation. The development of the process over large lengths of the contact device is investigated, and the asymptotic values of the component concentrations are determined.

Modern theory and industrial practice require adequate methods for investigating and calculating complex mass-exchange processes. The development of such methods, in turn, requires the tool of differential equations of convective mass transport [1]. When used for the description of multicomponent systems, these equations are characterized by matrix vectors and a conjugate form, which makes their solution difficult. For two-phase, multicomponent systems, the solution becomes considerably more complex, since the matrix conjugation equations which relate the component concentrations of both phases at the interface also have a conjugate character.

The present article presents a theoretical investigation of the mass-exchange process in a multicomponent, gas-liquid mixture, based on solving the system of differential equations of multicomponent convective diffusion in conjugate form.

Consider an n-component mixture moving in a rectangular channel as a descending, straight-through flow of phases. The x axis is oriented along the channel axis, while the y axis is perpendicular to it (Fig. 1). We make the usual assumptions [2-4]: The physical characteristics of the phases are constant, external forces are absent, phase equilibrium conditions prevail at the interface, and the thickness of the liquid film is constant. Then, the equations of convective, multicomponent diffusion are given by

$$u_{\mathbf{g}}(y)\frac{\partial \mathbf{C}_{\mathbf{g}}}{\partial x} = [D_{\mathbf{g}}]\frac{\partial^2 \mathbf{C}_{\mathbf{g}}}{\partial y^2}.$$
 (1)

$$u_{\mathbf{g}}(y) \frac{\partial \mathbf{C}_{\mathbf{g}}}{\partial x} = [D_{\mathbf{g}}] \frac{\partial^2 \mathbf{C}_{\mathbf{g}}}{\partial y^2}, \qquad (2)$$

while the boundary conditions are assigned by the following relationships:

at the channel inlet,

 $x = 0, \quad \mathbf{C}_{\mathbf{g}} = \mathbf{C}_{\mathbf{0}\mathbf{g}}, \quad \mathbf{C}_{\mathbf{g}} = \mathbf{C}_{\mathbf{0}\mathbf{g}};$ (3)

at the channel wall (impenetrability condition),

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