LITERATURE CITED

1. L. N. Sretenskii, The Theory of Wave Motions of a Fluid [in Russian], Nauka, Moscow (1977).
2. N. Salvesen, "On higher-order wave theory for submerged two-dimensional bodies," J. Fluid Mech., 38, 415 (1969).
3. V. I. Bukreev, "Waves generated at an interface between two fluids of different densities by the motion of a circular cylinder and a symmetric wing," Zh. Prikl. Mekh. Tekh. Fiz., No. 1 (1980).
4. N. N. Borodina, V. I. Bukreev, A. V. Gusev, and I. V. Sturova, "Viscous damping of internal waves generated in a two-layer liquid by the motion of a cylinder and wing," in: Mathematical Problems of the Mechanics of Continuous Media [in Russian], No. 54, Inst. Hydrodynamics, Siberian Branch, Academy of Sciences of the USSR, Novosibirsk (1982).

DIFFUSION OF A SPARINGLY SOLUBLE GAS IN A FLOWING LIQUID FILM

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A film of liquid flowing over an inclined surface is almost always covered by waves, which have a substantial effect on the transfer between phases. The effect of the waves is most pronounced when a sparingly soluble gas is taken up by the film. The increase in the mass-transfer coefficient caused by the waves can be 100% or more [1, 2].

Several factors have been described as responsible for increasing the mass transfer in waves: increase in the overall phase interface area due to the waves (not more than 1% [1]), destruction of the inert gas layer hindering absorption by the waves (although this does not hinder desorption [1]), vortex motion in the gas and liquid phases [2], the presence of vigorous convective flows [3, 4], and complete mixing in the wave depressions [5].

The main resistance to diffusion is localized in the liquid if the gas is sparingly soluble, so diffusion in the gas can be neglected. No vortex motion has been observed up to Reynolds numbers of Re = 200-400 in various experiments on visualizing the flow in films [6, 7], so the intensification is due purely to the wave mechanism [1].

Here we used the thin boundary-layer approximation near the free surface to obtain an exact solution for the diffusion in such a film. It is shown that the main factors that increase the mass transfer for two-dimensional waves are firstly that the surface velocity of the liquid at the wave crests approximates to the phase velocity, which leads to the interesting phenomenon of diffusion independence in the cells of liquid separated by the ridges, and, secondly, the vigorous transverse velocity fluctuations.

Hydrodynamics of a Wave Film. When there are fluctuations in flow rate, one gets two-dimensional waves [1, 8]. The experiments of [1, 8] imply that the wavelength greatly exceeds the average film thickness over a large range in flow rate. The longitudinal-velocity profile is closely approximated by a polynomial of second degree [6]:

$$u(x, y, t) = \frac{3}{2} V(x, t)\left[2\frac{y}{h(x, t)} - \left(\frac{y}{h(x, t)}\right)^2\right], \tag{1}$$

where V is the film velocity averaged over the cross section; y, normal coordinate reckoned from the wall to the free surface; x, longitudinal coordinate; t, time; and h, instantaneous film thickness.

We take the waves as stationary [i.e., $h = h(x - ct)$, where c is the phase velocity] which gives us $hV = hc + \text{const}$ from the condition for constancy of the flow rate in the wave system [9]. We average this over the wavelength to determine the constant as $q - \langle h\rangle c$, where q is the average flow rate in the laboratory coordinate system and $\langle h\rangle$ is the average film thickness. We get for the velocity averaged over the cross section that

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$$V = c - (c\langle h\rangle - q)/h. \tag{2}$$

With a given flow rate q, the film thickness is determined from the balance between the force of gravity and friction averaged over the wavelength [the latter calculated from (1)]:

$$\frac{1}{\lambda}\int_0^\lambda gh\,d\xi = \frac{1}{\lambda}\int_0^\lambda 3\nu\frac{V}{h}\,d\xi. \tag{3}$$

We introduce dimensionless variables: velocity $V' = V\langle h\rangle/q$, local film thickness $h' = h/\langle h\rangle$, the Reynolds number $\mathrm{Re} = q/\nu$, and the wave phase $\xi' = (x - ct)/\lambda$, when (3) becomes

$$\frac{\langle h\rangle^3 g}{q^2} = \frac{3}{\mathrm{Re}}\int_0^1 \frac{V'}{h'}\,d\xi'.$$

From this, on the basis of (2), we get a formula for the ratio of the average thicknesses of the film with waves $\langle h\rangle$ and without waves h_0 for a given flow rate (Re = idem):

$$\frac{\langle h\rangle}{h_0} = \left(\int_0^1 \frac{c'(h'-1)+1}{h'^2}\,d\xi'\right)^{1/3}, \tag{4}$$

where $c' = c\langle h\rangle/q$ is the dimensionless phase velocity.

The dimensionless velocity of the liquid at the surface of the film is given by

$$w' = \frac{3}{2}V' = \frac{3}{2}\left(c' - \frac{c'-1}{h'}\right).$$

We integrate the equation of continuity to get the normal component of the velocity near the free surface as

$$v = v_h - \int_h^y \frac{\partial u}{\partial x}\,dy \cong v_h - (y-h)\frac{\partial w}{\partial x},$$

where v_h is the normal velocity of the film surface, which is related to the surface velocity w by the kinetic condition

$$v_h = \partial h/\partial t + w\,\partial h/\partial x. \tag{5}$$

In what follows we use the following dimensionless variables: $w' = w\langle h\rangle/q$, $v' = v\lambda/q$, $x' = x/\lambda$, $y' = y/\langle h\rangle$, $t' = tq/\lambda\langle h\rangle$, $\varepsilon = \langle h\rangle/\lambda \ll 1$.

Diffusion in a Wave Film. The diffusion coefficients for gases in liquids are usually very small ($D \sim 10^{-10}$ m^2/sec). This means that even very long parts of the film can be considered in the thin boundary-layer approximation for the free surface. In that approximation, the dimensionless diffusion equation takes the form

$$\frac{\partial\theta}{\partial t} + w\frac{\partial\theta}{\partial x} + \left(v_h + (h-y)\frac{\partial w}{\partial x}\right)\frac{\partial\theta}{\partial y} = \frac{1}{\mathrm{Pe}\,\varepsilon}\frac{\partial^2\theta}{\partial y^2}, \tag{6}$$

where $\mathrm{Pe} = q/D$ is the Peclet number; $\theta = (C - C_h)/(C_0 - C_h)$, dimensionless concentration; and C_0 and C_h, initial concentration (at x = 0) and the surface one (at y = h). Here and subsequently, we omit the primes with the dimensionless quantities. The following are the boundary and initial conditions for (6):

$$\theta(y = h) = 0,\quad \theta(x = 0) = \theta(y = \infty) = 1. \tag{7}$$

For a stationary wave we have h(x − ct) and w(x − ct), so it is convenient to convert to the new variables

$$x,\quad \xi = x - ct,\quad z = h - y. \tag{8}$$

We use condition (7) with the variables of (8) to put (6) in the form

$$w\frac{\partial\theta}{\partial x} + (w-c)\frac{\partial\theta}{\partial\xi} - z\frac{dw}{d\xi}\frac{\partial\theta}{\partial z} = \frac{1}{\mathrm{Pe}\,\varepsilon}\frac{\partial^2\theta}{\partial z^2}.$$

We assume that the concentration θ is dependent only on one self-modeling variable $\eta = z/\delta(x, \xi)$, where δ is some nominal thickness for the diffusion layer. The equations for $\theta(\eta)$ and $\delta(x, \xi)$ separate:

$$\begin{gathered} -2\eta d\theta/d\eta = d^2\theta/d\eta^2; \\ w\frac{\partial\delta^2}{\partial x} + (w-c)\frac{\partial\delta^2}{\partial\xi} + 2\frac{dw}{d\xi}\delta^2 = \frac{4}{\mathrm{Pe}\,\varepsilon}. \end{gathered} \tag{9}$$

The solution for θ that satisfies the boundary conditions of (7) is written as

$$\theta = \frac{2}{\sqrt{\pi}}\int_0^\eta e^{-\eta^2}d\eta. \tag{10}$$

We determine δ^2 by the characteristic method. The characteristic equation in the x,ξ plane

$$dx/d\xi = w/(w-c)$$

gives us the family of curves

$$x = \gamma + \int_0^\xi \frac{w}{w-c}d\xi, \tag{11}$$

dependent on parameter γ. Along the curve of (11), the first term in (9) takes the form $(w-c)\frac{dx}{d\xi}\frac{\partial\delta^2}{\partial x}$; when we have multiplied all terms in (9) by $w - c$, we transform the left side to the total derivative with respect to ξ. We integrate with respect to ξ to get

$$(w-c)^2\delta^2 = \frac{4}{\mathrm{Pe}\,\varepsilon}\int_0^\xi (w-c)\,d\xi + A(\gamma). \tag{12}$$

The constant of integration A is dependent on parameter γ, which defines the characteristic curve of (11). This relationship is obtained in parametric form from (11) and (12) with the initial condition $\delta(x = 0) = 0$:

$$A(\gamma) = \frac{4}{\mathrm{Pe}\,\varepsilon}\int_0^\xi (c-w)\,d\xi; \tag{13}$$

$$\gamma = f(\xi) = \int_0^\xi \frac{w}{c-w}d\xi. \tag{14}$$

The function $f(\gamma)$ is defined by the integral of (14) and is monotone in $c > w > 0$, so we introduce the inverse function $\xi = p(\gamma) = f^{-1}(\gamma)$. We substitute $p(\gamma)$ into the upper limit in (13) and substitute (13) into (12); we determine γ from (11) and after obvious steps have

$$\delta^2 = \frac{4}{\mathrm{Pe}\,\varepsilon\,(c-w)^2}\int_\xi^{p(x+f(\xi))} (c-w)\,d\xi. \tag{15}$$

The expression for the dimensionless local mass flux follows from the solutions (10) and (15):

$$\left(\frac{\partial\theta}{\partial z}\right)_{z=0} = \frac{2}{\sqrt{\pi}\,\delta} = (c-w)\left[\frac{\pi}{\mathrm{Pe}\,\varepsilon}\int_\xi^{p(x+f(\xi))} (c-w)\,d\xi\right]^{-1/2}. \tag{16}$$

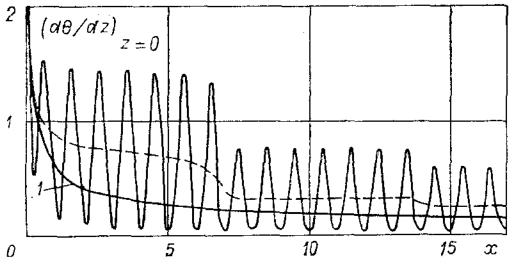

Fig. 1

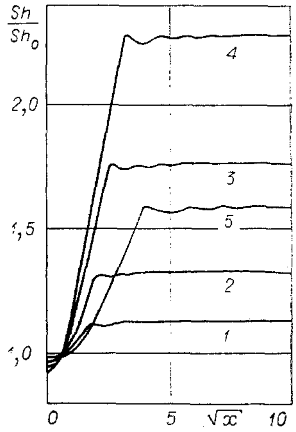

Fig. 2

We integrate (16) with respect to x and average with respect to ξ from 0 to 1 to get the integral mass flux, which is usually monitored in experiments from the difference in the concentrations at the film inlet and outlet [1]. However, the integration with respect to x can be avoided if we bear in mind that (9) can be written as

$$\frac{2}{\sqrt{\pi\varepsilon\delta}} = \frac{\mathrm{Pe}}{\sqrt{\pi}}\left\{\frac{\partial}{\partial x}(w\delta) + \frac{\partial}{\partial \xi}[(w-c)\delta]\right\}. \tag{17}$$

After integrating (17) with respect to x and averaging with respect to ξ, we get the dimensionless total mass flux, which is known in the literature as the Sherwood criterion [1]:

$$\mathrm{Sh} = \int_0^x dx \int_0^1 \frac{2}{\sqrt{\pi\varepsilon\delta}}\, d\xi = \frac{\mathrm{Pe}}{\sqrt{\pi}} \int_0^1 w\delta\, d\xi.$$

The effects of the waves on the mass transfer are naturally evaluated from the ratio of the total mass fluxes for films with and without waves:

$$\frac{\mathrm{Sh}}{\mathrm{Sh}_0} = \left(\frac{3\langle h\rangle}{2h_0}x\right)^{-1/2} \int_0^1 \left[\int_\xi^{p(x+f(\xi))} (c-w)\, d\xi\right]^{1/2} \frac{w\, d\xi}{c-w}. \tag{18}$$

From (18) we can obtain certain general results unrelated to the particular form of $w(\xi)$. At equally separated points $x_n = nf(1)$ $(n = 1, 2, \ldots)$, the ratio of (18) takes identical values. In fact, at these points the upper limit to the integral in (18) is $p(nf(1) + f(\xi)) = p(f(n + \xi)) = n + \xi$. The inner integral in (18) is calculated as $\int_0^1 (c-w)\, d\xi$, while the outer one is expressed in terms of f(1); as a result, we have

$$\left(\frac{\mathrm{Sh}}{\mathrm{Sh}_0}\right)_{\max} = \left[\frac{2h_0}{3\langle h\rangle} f(1) \int_0^1 (c-w)\, d\xi\right]^{1/2}. \tag{19}$$

It can be shown that (19) defines the maxima in $\mathrm{Sh}/\mathrm{Sh}_0$.

Note that the ratio of (19) attains arbitrary large values when $w(\xi)$ attains the phase velocity c at a certain point. Then the f(1) appearing in (19) becomes infinite in accordance with (14).

For x small, the upper limit in the inner integral in (18) is expanded as a Taylor series, from which we take only two terms, and we have for $x \to 0$ that $p(x + f(\xi)) \cong \xi + x/f'(\xi) = \xi + (c - w)x/w$, which gives

$$\left(\frac{\mathrm{Sh}}{\mathrm{Sh}_0}\right)_{x=0} = \left(\frac{2h_0}{3\langle h\rangle}\right)^{1/2} \int_0^1 w^{1/2}\, d\xi. \tag{20}$$

From the latter expression one can obtain values somewhat less than one.

Calculation for a Harmonic Wave. Qualitative Interpretation. In what follows we put everywhere that $h = 1 + \alpha \cos 2\pi\xi$, where α is the relative wave amplitude. The integral in (4) is calculated by differentiation with respect to the parameter, which gives

$$\langle h \rangle / h_0 = (1 - ca^2)^{1/3} / (1 - a^2)^{1/2}.$$

Similarly, we calculate the other integrals:

$$f(1) = \frac{6(c-1)}{\sqrt{(2c-3)^2 - c^2a^2}} - 3, \quad \int_0^1 (c - w)\, d\xi = \frac{3(c-1)}{2\sqrt{1-a^2}} - \frac{c}{2}$$

appearing in (19). Formula (20) is expressed in a complicated way in terms of elliptic integrals and is not given here.

The function f and the inverse one p are monotonically increasing functions and pass through the origin. The integral defining $f(\xi)$ in accordance with (14) and all the other integrals in (16) and (18) are calculated by Simpson's method.

Figure 1 shows the instantaneous local mass flux at the surface on an arbitrary scale along the vertical as calculated for $c = 2$ and $\alpha = 0.4$ from (16), and it also gives curve 1 related to the wave-free case ($\alpha = 0$). We have taken an instant where the wave crests lie at points with integer values of the coordinate x. It is evident that the minima in the local mass flux occur near these points, while the maxima lie in the wave depressions. There is a certain displacement of the extremal points in the positive direction near the origin.

Figure 2 shows the dependence on $\sqrt{x}$ for the ratio Sh/Sh_0 calculated from (18): Curves 1-4 have been calculated for $c = 2$ and $\alpha = 0.2$, 0.3, 0.4, and 0.45, correspondingly, while curve 5 is for $c = 1.7$ and $\alpha = 0.2$. The heights of the peaks correspond precisely to (19), and they lie at the points $x_n = nf(1)$, which are the points where there is a sharp reduction in the fluctuation level for the local mass flux in Fig. 1.

The following is a qualitative explanation for this behavior of the curves. We transfer to the wave system, where the surface velocity is $c - w$ and is directed toward negative x. The current lines come together in the depressions and expand at the crests. The thickness δ of the diffusion layer decreases and the local flux is maximal at points where the liquid flows toward the free surface. This explains the positive shift in the turning points in Fig. 1 at small x, where diffusion in the wave system has not yet stabilized and the initial condition makes itself felt.

The time to transport any part of the liquid along the surface by one wavelength is determined by the integral $\int_0^1 d\xi/(c - w)$; if one follows an individual wave cell, then at this instant the trough receives liquid with an elevated concentration from an adjacent trough, which should produce a sharp fall in the local mass flux (Fig. 1). This first occurs at the distance defined by

$$c \int_0^1 \frac{d\xi}{c - w} - 1 = \int_0^1 \frac{w\,d\xi}{c - w} = x_1. \tag{21}$$

The one in this expression is subtracted because there is a shift of one wavelength in the negative sense during the time of drift from cell to cell. The larger the drift time, the longer in x of a part in which the diffusion in the cells stabilizes (linear growth parts in Fig. 2). If the maximum value $w = c$ is attained somewhere on the surface, the integral in (21) diverges ($x_1 \to \infty$) and the entire film will consist of cells separated by wave crests that are indendent of one another in a diffusion respect. The integral mass flux will then clearly be proportional to x, in contrast to the square-root law for a wave-free film ($Sh_0 \sim \sqrt{x}$). High values of Sh/Sh_0 obtained also for waves of small amplitude but close to the critical value $\alpha_* = 2 - 3/c$ derived from the condition $w = c$ (for example, curve 5 in Fig. 2). However, the mass-transfer rate for small amplitudes is less than that for large ones (small slope of the linear part of curve 5 in Fig. 2).

The time-averaged local flux will have almost a stepped form (broken line in Fig. 1) and, therefore, the integral flux will be almost piecewise-linear with kinks at the points $x_n = nf(1)$: In the wave system, these points correspond to successive instants when the depression receives parts of the liquid that at t = 0 were in the depressions in the successively more remote cells. The piecewise-linear character of Sh(x) explains the behavior of the curves in Fig. 2 (increase linear in $\sqrt{x}$ for Sh/Sh_0 in the initial part, oscillations, and asymptote).

Therefore, the intensification of the mass transfer in a wave cell is determined by two basic factors: by the amplitude and by the closeness of the liquid speed at the crests to the phase velocity. The transverse velocity fluctuations increase with the amplitude, which reduce the thickness of the diffusion layer in the depressions and markedly increase the local mass flux in these parts. With a given amplitude, any reduction in the phase velocity produces a reduction in the critical value a_* and at a certain $c = c_*$ we get $a = a_*$, and at the crests the liquid overtakes the waves and the cells become diffusion-independent. At large distances, the value of Sh/Sh_0 in that case can be arbitrarily large.

These conclusions evidently apply for any two-dimensional wave, and in particular for the so-called large-amplitude rolling waves, at the crests of which one finds $w \approx c$ [6]. The destruction of the rolling waves, which give way to three-dimensional nonstationary formations, should substantially reduce the mass transfer, since the isolated wave cells are eliminated. This is observed in experiments on the artificial disruption of two-dimensional waves [1].

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LITERATURE CITED

1. V. E. Nakaryakov, B. G. Pokusaev, and S. V. Alekseenko, "Desorption of a sparingly soluble gas from flowing wave films of liquid," in: Calculation on Heat and Mass Transfer in High-Energy Chemical Processes [in Russian], A. P. Burdukov (ed.), ITF Sib. Otd. Akad. Nauk SSSR, Novosibirsk (1981).
2. R. A. Seban, "Transport to falling films," in: Proc. 6th Int. Heat Transfer Conf., Vol. 6, Ottawa (1979).
3. S. D. Bagaeva, P. A. Semenov, and M. F. Galiullin, "Diffusion in the wave motion of a thin layer of liquid," TOKhT, 7, No. 4 (1972).
4. I. P. Semenova, Acceleration of Mass Transfer by Waves at the Phase Interface in Separate Flow in a Gas–Liquid Mixture [in Russian], Izd. NII Mekh. Mosk. Gos. Univ. (1978).
5. L. P. Kholpanov, V. Ya. Shkadov, et al., "The mass transfer in a film of liquid when waves are formed," TOKhT, 1, No. 1 (1967).
6. V. E. Nakoryakov, B. G. Pokusaev, et al., "The instantaneous velocity profile in a wave film of liquid," Inzh.-Fiz. Zh., 33, No. 3 (1977).
7. F. Ho and R. Hummel, "Average velocity distributions within falling liquid films," CES, 25, 1225 (1970).
8. S. V. Alekseenko, V. E. Nakoryakov, and B. G. Pokusaev, Waves at the Surface of a Vertically Flowing Liquid Film, Preprint 36-79 [in Russian], ITF Sib. Otd. Akad. Nauk SSSR, Novosibirsk (1979).
9. P. L. Kapitsa, "Wave flow in thin layers of viscous liquid," Zh. Eksp. Teor. Fiz., 18, No. 3 (1948).