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MASS TRANSFER TO FALLING LIQUID FILMS AT LOW REYNOLDS NUMBERS

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Abstract—A non-linear treatment of the hydrodynamics of wave motion is suggested and its results are used for the solution of the mass-transfer problem. An equation of convective-diffusion valid in the small penetration case is established and is solved exactly by means of a similarity transformation.

The main result obtained is that for low Reynolds numbers, the average amplification factor $\bar{\alpha}$ (the time average of the ratio between the mass flux in wave motion and in laminar steady motion) is a function, having the form from Fig. 4, only upon the dimensionless quantity $\psi = g^{1/6}Q^{11/6}/\nu^{7/6}(\sigma/\rho)^{1/2}$.

NOMENCLATURE

a, $= |2i\varphi_1|$;
a_n, coefficients in the expansion (1);
A_q, B_q, C_q, D_q, dimensionless coefficients in the expansions (3);
$A_{q,2r}, B_{q,2r}, C_{q,2r}, D_{q,2r}$, coefficients in the expansions of $A_q, \ldots, D_q$ with respect to φ_1;
c, concentration;
c_0, value of c for $y_1 = 0$;
d, quantity defined by equation (A-7);
D, diffusion coefficient;
g, acceleration of gravity;
h, film thickness;
h_0, average film thickness;
H_0, film thickness in the laminar steady case;
H_{2r}, coefficients in the expansion of h_0 with respect to φ_1;
$I_1(z), I_2(z)$, functions defined by equations (29) and 27);
k, wave number;
k_{2r}, coefficients in the expansion of k with respect to φ_1;
N, mass flux at the free interface;
N_l, mass flux at the free interface in the case of a steady laminar motion;
Q, average liquid flow rate [cm^3/cm s];
t, time;
u, x component of the liquid velocity;
u_0, value of u for $y_1 = 0$;
v, y component of velocity;
v_1, y_1 component of velocity;
w, wave velocity;
x, distance along the wall;
x_1, $= kx$;
x_1', quantity defined by equation (A-6);
y, distance to the wall;
y_1, distance to the free interface ($y_1 \approx h - y$);
z, $= k(x - wt)$.

Greek symbols

α, amplification factor defined by equation (32);
$\beta_0, \beta_1, \beta_2, \beta_3, \ldots, \beta_6$, coefficients defined by equation (9);
δ, thickness which appears in the dimensionless variable η;

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Δ, quantity defined by equation (25);
ε^2, arbitrary numerical constant;
η, $= y_1/\delta$;
χ, $= \dfrac{wh_0}{Q}$;
χ_{2n}, coefficients in the expansion of χ with respect to φ_1;
φ, ratio between elongation and average film thickness $\left(\varphi = \dfrac{h - h_0}{h_0}\right)$;
φ_1, coefficient of e^{iz} in the Fourier expansion of φ;
ν, kinematic viscosity of the liquid;
ψ, dimensionless parameter defined by equation (8);
ρ, density of liquid;
σ, surface tension;
θ, quantity defined by equation (27).

Subscripts
n, index in the expansion with respect to y_1 [equation (1)];
q, index in the Fourier expansion [equation (3)];
r, index in the expansion with respect to φ_1 [equation (7)].

Superscripts
$\bar{}$, temporal average,
$\hat{}$, real part of a complex quantity;
$\sim$, imaginary part of a complex quantity.

INTRODUCTION

FRIEDMAN and Miller [1] and, since then, many other investigators have observed that when a thin layer of liquid flows along a smooth vertical plate, its free surface is not plane but is disturbed by wave motions and ripples. It is therefore natural to attribute to these unsteady motions the fact that the rate of absorption in a falling liquid film is up to 200 per cent larger [2–5] than that predicted by the equation valid for steady laminar motion.

The hydrodynamical problem was examined from the point of view of the linear instability theory by Yih [6] and by Benjamin [7] who have shown that Nusselt's velocity distribution is not stable to small perturbations. This approach cannot lead, however, to information concerning the stable unsteady velocity distribution. As an approximation one can consider as significant for the stable unsteady state, the dominant wave length (i.e. the wave length for which the real part of the growth constant which multiply the time in the expression of the perturbation has a maximum) and obtain in this manner equations for the wave length and for the wave velocity. Kapitza [8] has suggested previously another approach based on the assumption that the film thickness and the velocity components are periodical functions of $z = k(x - wt)$. Using for u an equation of the Nusselt type, he obtained for φ an approximate equation to which the condition of periodicity is imposed. One obtains in this manner equations for the wave length, wave velocity and velocity distribution. One may note that the equations obtained by Kapitza for the wave length and wave velocity do not differ from the ones obtained by the approximate procedure mentioned above. This is not surprising, since their establishment in [8] imply in fact a kind of linearization of the equations of motion. The equations obtained lead for the film thickness, the wave length and wave velocity to values of the same order of magnitude as the experimental ones [9–11]. However, Kapitza's theory predicts a strong dependence of the wave length on velocity, while experiment shows it to be practically independent on velocity; it predicts also a constant value for the ratio between the wave velocity and the average liquid velocity, while experiment shows a decrease with the increase of the average velocity of the liquid.

On the basis of the velocity distribution obtained by Kapitza one can solve the convective diffusion equation. The problem was first examined by Levich [12] who writes the concentration as a sum of two terms $(c_1 + c_2)$; the first c_1 represents the concentration in the case of a laminar steady motion and the second c_2 the correction due to wave motion. Introducing this expression in the convective-diffusion equation and averaging with respect to time, he obtains an equation for the average value of the correction. That equation is not, however, correct, since the average value of $v(\partial c_2/\partial y)$ is not zero as it is assumed in its deduction. Levich's method of calculation depends essentially upon the possibility of ignoring this term and cannot be, for this reason, corrected. The problem was solved by Ruckenstein and Berbente [13] who have shown that the use of Kapitza velocity distribution leads to the conclusion, which disagrees with experiment, that the rate of mass transfer is only increased 30 per cent by the wave motion. It was, however, noticed that the agreement may be much improved if one uses in Kapitza's velocity distribution the experimental values for the amplitude, wave length and wave velocity, instead of the theoretical ones.

A new theoretical approach is therefore needed for the hydrodynamics of wave motion. An attempt in this direction was made by Berbente and Ruckenstein [14] who, unlike the theories mentioned above, have given a nonlinear treatment of the hydrodynamic problem, by looking for periodical solutions of z of the non-linear equations of motion. It is the aim of the present paper to review briefly (without mathematical details) that approach and to use its results in the solution of the mass-transfer problem.

The main result obtained here is that in the range of validity of the thin film approximation used in the treatment of the hydrodynamic problem (low Reynolds numbers), the average amplification factor $\bar{\alpha}$ is a function only of the dimensionless quantity

$$\psi = \frac{g^{1/6} Q^{11/6}}{\nu^{7/6} (\sigma/\rho)^{1/2}}.$$

The form of the dependence is obtained theoretically. The results obtained are in qualitative and quantitative agreement with experiment.

HYDRODYNAMICS OF WAVE FLOW

In this section we shall review the method developed by the authors for obtaining periodical solutions of the non-linear equations of motion. Though it may be applied to any value of Reynolds number, the calculations were carried out, for simplicity's sake, only in the "thin film approximation". In the thin film approximation the y component of velocity is considered small (consequently the pressure does not depend upon y) and $|\partial^2 u/\partial x^2| \ll |\partial^2 u/\partial y^2|$. We notice that there exists similarities between the hydrodynamic equations used in the thin layer approximation and those used in the boundary-layer approximation.

The method consists of three steps:

The first step is an expansion with respect to y_1 of the form

$$u = \frac{3}{2} \frac{Q}{h_0} \sum_{n=0}^{\infty} g_n \left(\frac{y_1}{h_0} \right)^n. \tag{1}$$

This expansion was restricted to the terms up to the sixth degree with respect to y_1/h_0. Introducing expansion (1) in the Navier–Stokes equations, written in the thin film approximation, and using the boundary conditions one obtains $a_1 = a_3 = a_5 = 0$ and a number of five nonlinear equations for a_0, a_2, a_4, a_6 and φ which are considered as functions only of z.

$$z = k(x - wt). \tag{2}$$

In the second step one imposes to a_0, a_2, a_4, a_6 and φ the condition to be periodical functions of z and one uses for them Fourier series expansions of the form

$$\left.\begin{aligned} a_0 &= 1 + \sum_{-\infty}^{\infty} A_q e^{iqz}, \\ a_2 &= -1 + \sum_{-\infty}^{\infty} B_q e^{iqz} \end{aligned}\right\} \tag{3}$$

$$\left.\begin{aligned} a_4 &= \sum_{-\infty}^{\infty} C_q \, e^{iqz}, \\ a_6 &= \sum_{-\infty}^{\infty} D_q \, e^{iqz}, \\ \varphi &= \sum_{-\infty}^{\infty} \varphi_q \, e^{iqz}. \end{aligned}\right\} \quad (3)$$

In these expansions A_q, B_q, C_q, D_q and φ_q are constants. The first term in the expansions of a_0 and a_2 represents their values for the case of a steady laminar motion. The values of a_4, a_6 and φ are zero in the last case. Since the equations for $a_0, \ldots, \varphi$ are non-linear, the resulting equations for $A_q, \ldots, \varphi_q$ are also non-linear. The number of unknown quantities is larger than the number of equations, the difference being equal to two. Since the periodical quantities are determined only up to a phase constant, one may choose for this arbitrary phase constant such a value that the calculations be simplified. For this reason we consider in the expansion of φ that the coefficient of cos z is nil, i.e.

$$\varphi_1 + \varphi_{-1} = 0. \quad (4)$$

A single quantity remains undetermined. We selected φ_1 as that quantity since it has a simple physical meaning, being proportional with the first approximation of the wave amplitude. Indeed, in the first approximation

$$\varphi = \varphi_1 \, e^{iz} + \varphi_{-1} \, e^{-iz} \quad (5)$$

which, taking (4) into account, leads to

$$\varphi = 2i\varphi_1 \sin z. \quad (6)$$

Therefore all the constants A_q, B_q, C_q, D_q, φ_q may be obtained as functions of φ_1.

The third step consists in the expansion of these constants with respect to φ_1. One may prove that the expansions have the forms

$$h_0 = \sum_{r=0}^{\infty} H_{2r} \varphi_1^{2r}, \quad \chi = \sum_{r=0}^{\infty} \chi_{2r} \varphi_1^{2r}, \quad k = \sum_{r=0}^{\infty} k_{2r} \varphi_1^{2r},$$
$$A_q = \varphi_1^{|q|} \sum_{r=0}^{\infty} A_{q,2r} \varphi_1^{2r}, \ldots, \varphi_q = \varphi_1^{|q|} \sum_{r=0}^{\infty} \varphi_{q,2r} \varphi_1^{2r}. \quad (7)$$

The coefficients $A_{q,2r}, \ldots$ may be calculated by introducing expansions (7) into the system of equations obtained for $A_q, \ldots$ and by identifying the terms in $\varphi_1^{|q|+2r}$. One obtains a system having an infinite number of equations which is, however, formed by a succession of systems of equations each containing five equations with five unknown complex quantities. For this reason, the system of equations may be solved easily, since the unknown quantities result step by step, in increasing order of the powers of φ_1. We note also the advantage that each system of five equations is linear with respect to the quantities which must be determined in that step, the non-linear terms in these equations containing only quantities calculated in the preceding steps.

The calculations lead to the conclusion that in the framework of the thin film approximation all the dimensionless coefficients

$$\frac{H_{2r}}{H_0}, \frac{k_{2r}}{k_0}, A_{q,2r}, B_{q,2r}, C_{q,2r}, D_{q,2r}, \varphi_{q,2r}$$

are functions of a single dimensionless parameter

$$\psi = \frac{g^{\frac{1}{3}} Q^{\frac{11}{9}}}{\nu^{\frac{4}{3}} (\sigma/\rho)^{\frac{1}{3}}}. \quad (8)$$

Our theoretical approach cannot provide information concerning φ_1. A similar difficulty exists also in Kapitza's theory. This author has used the condition of minimum dissipation in order to determine the value of the amplitude. Such an extremum condition may be imposed also to other physical quantities as are, for instance, h_0, k, the average value of the free surface area. The values for the amplitude obtained by us in this manner differ from one another. Nevertheless, we remark that whatever is the "correct" physical quantity chosen for the determination of φ_1 by means of an extremum principle it may be written, as mentioned above, in the form

$$\bar{f} = f_0(1 + f_2(\psi)\,\varphi_1^2 + f_4(\psi)\,\varphi_1^4 + \ldots).$$

The extremum condition leads to the conclusion that φ_1 is a function of ψ. The form of the function was determined by using Kapitza's experimental values for the amplitude. As results from Fig. 1, the experimental values obtained by Kapitza for water and ethyl–alcohol are well fitted by a single curve.

One may, therefore, conclude that the wave motion is characterized in the range of validity of the thin film approximation by a single dimensionless group ψ. We notice that outside this range, besides ψ the wave motion is characterized also by the Reynolds number.

The evaluations made (see [14]) show that the thin film approximation is valid if $kh_0 \leqslant 0{\cdot}30$. On the other hand the restriction of the expansion up to the term of sixth degree in y_1/h_0 may be made if $\psi \leqslant 5$. For water the two inequalities are satisfied if $Re < 100$.* In the range of validity of the mentioned restrictions the amplitude is small and so the calculation may be performed only up to the third order approximation (inclusively) with respect to φ_1.

In order to predict the rate of mass transfer we need to know (see the following section of the paper) a_0, χ, k and h_0.

For a_0 one obtains the equation

$$a_0 = 1 - \frac{a^2}{4} A_{02} - a\left(\hat{A}_{10} - \frac{a^2}{4}\hat{A}_{12}\right)\sin z - a\left(\tilde{A}_{10} - \frac{a^2}{4}\tilde{A}_{12}\right)\cos z$$
$$- \frac{a^2}{2}(\hat{A}_{20}\cos 2z - \tilde{A}_{20}\sin 2z) + \frac{a^3}{4}(\hat{A}_{30}\sin 3z + \tilde{A}_{30}\cos 3z) \equiv \beta_0 + \beta_1 \sin z$$
$$+ \beta_2\cos z + \beta_3 \sin 2z + \beta_4\cos 2z + \beta_5 \sin 3z + \beta_6\cos 3z \qquad (9)$$

where $a = |2i\varphi_1|$.

In Fig. 2 $\beta_0, \beta_1, \beta_2, \beta_3, \ldots, \beta_6$ are plotted as functions of ψ.

In Fig. 3 $\chi/3$, k/k_1 and h_0/H_0 are plotted as functions of ψ. For comparison experimental points are also given. There exists a qualitative and a quantitative agreement between theory and experiment.

The quantities k_1 and H_0 are given by the expressions:

$$k_1 = \left(1{\cdot}2\,\frac{g\rho Q}{\nu\sigma}\right)^{\frac{1}{2}} \qquad (10)$$

$$H_0 = \left(\frac{3\nu Q}{g}\right)^{\frac{1}{3}}. \qquad (11)$$

The wave motion is in reality more complex than described by our approach. Tailby and Portalski [10] and Stainthorp and Allen [11] have studied photographically the wave motion and noticed the following observations: (1) the inception of wave formation does not start at the leading edge of the wetted plate, but at some distance below it, depending, *inter alia*, on Reynolds number; (2) the wave length depends not only on the flow rate and on the physical properties of the liquid, but is also a very complicated function of the height of the wetted wall; (3) the wave motion loses its regularity some distance below the line of inception of wave motion. It appears from these observations that the proposed theoretical approach is valid for the region in the vicinity of the point of inception of wave motion. As a matter of fact, as stressed by Tailby and Portalski, this region of comparatively regular wave motion is the only one which may be used for determining experimentally wave lengths having the usual physical meaning.

* We notice that Kapitza's theory is also valid only for low Reynolds numbers.

RATE OF MASS TRANSFER

Since we are interested in calculating the

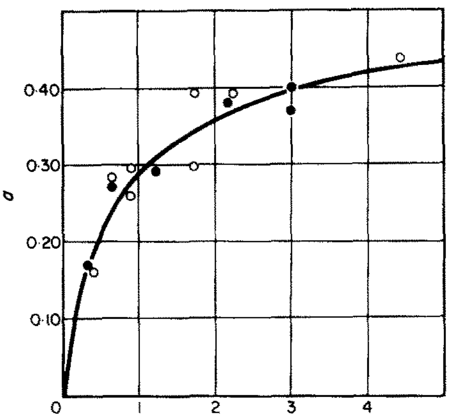

FIG. 1. a vs. ψ.
○ Kapitza's experimental data for water
● Kapitza's experimental data for ethyl alcohol

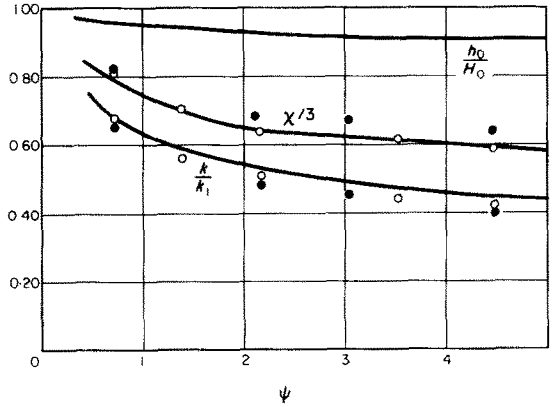

FIG. 3. $\chi/3$, k/k_1, h_0/H_0 vs. ψ.
— Author's theoretical curves
○ Kapitza's experimental data for water
● Kapitza's experimental data for ethyl alcohol

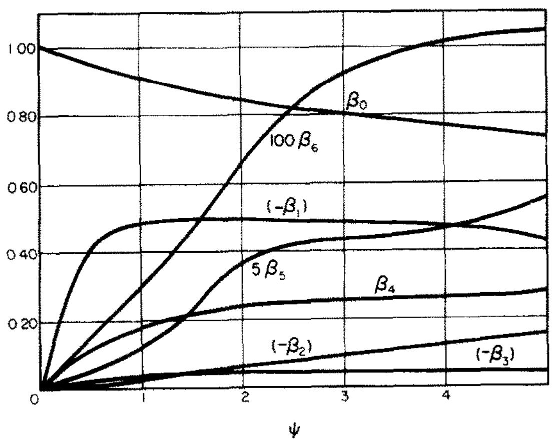

FIG. 2. Coefficients β_0, β_1, β_2, β_3, β_4, β_5, β_6 vs. ψ.

mass flux at the free surface it is natural to write the convective diffusion equation in a frame of reference bound to the free interface and to use the distance y_1 to the free surface and the distance x measured along the wall as the independent variables. This selection of the independent variables is convenient also owing to the fact that in cases where the depth of penetration by diffusion is small the concentration varies appreciably only in the vicinity of the interface.

The equation of convective-diffusion has the form

$$\frac{\partial c}{\partial t} + u\frac{\partial c}{\partial x} + v_1\frac{\partial c}{\partial y_1} = D\frac{\partial^2 c}{\partial y_1^2}, \tag{12}$$

where the velocity v_1 represents the velocity with respect to the free interface. Assuming that the depth of penetration by diffusion is small one may use for the velocity components expressions valid for small values of y_1. For small values of y_1 one may, however, write

$$u = u_0 + \left(\frac{\partial u}{\partial y_1}\right)_0 y_1 + \frac{1}{2}\left(\frac{\partial^2 u}{\partial y_1^2}\right)_0 y_1^2 + \ldots$$
$$\approx u_0 + \frac{1}{2}\left(\frac{\partial^2 u}{\partial y_1^2}\right)_0 y_1^2 \tag{13}$$

$$v_1 = v_{10} + \left(\frac{\partial v_1}{\partial y_1}\right)_0 y_1 + \ldots \approx \left(\frac{\partial v_1}{\partial y_1}\right)_0 y_1. \tag{14}$$

The evaluations made show that for the ranges of Reynolds number and ψ examined in the present paper one may neglect $\frac{1}{2}(\partial^2 u/\partial y_1^2)_0\, y_1^2$ as compared to u_0. We stress that for larger values of the Reynolds number and of the parameter ψ it seems that u_0 may become zero or possibly even negative for a part of the period so that it is no longer possible to neglect

$(\partial^2 u/\partial y_1^2)\, y_1^2$ as compared to u_0.* Consequently one can write for low Reynolds numbers

$$u \approx u_0 \tag{13a}$$

and

$$v_1 \approx \left(\frac{\partial v_1}{\partial y_1}\right)_0 y_1 \tag{14a}$$

and the equation of convective-diffusion may be transcribed under the form

$$\frac{\partial c}{\partial t} + u_0 \frac{\partial c}{\partial x} + \left(\frac{\partial v_1}{\partial y_1}\right)_0 y_1 \frac{\partial c}{\partial y_1} = D \frac{\partial^2 c}{\partial y_1^2}. \tag{15}$$

Since from the continuity equation and equation (1) we have

$$\left(\frac{\partial v_1}{\partial y_1}\right)_0 = -\left(\frac{\partial u}{\partial x}\right)_0 = -\frac{3}{2}\frac{Q}{h_0}\frac{\partial a_0}{\partial x} \tag{16}$$

equation (15) becomes, after the change of variable $z = x_1 - kwt$ is also performed,

$$-\left(\frac{2}{3}\chi - a_0\right)\frac{\partial c}{\partial z} + a_0 \frac{\partial c}{\partial x_1} - y_1 \frac{\mathrm{d}a_0}{\mathrm{d}z}\frac{\partial c}{\partial y_1} = \frac{2}{3}\frac{Dh_0}{Qk}\frac{\partial^2 c}{\partial y_1^2} \tag{17}$$

where a_0 is given by equation (9), χ, h_0 and k result from Fig. 3 and

$$x_1 = kx. \tag{18}$$

Equation (17) must be solved for the boundary conditions

$$c = c_0 \quad \text{for} \quad y_1 = 0 \tag{19a}$$

$$c = 0 \quad \text{for} \quad x = 0 \tag{19b}$$

$$c = 0 \quad \text{for} \quad y_1 \to \infty. \tag{19c}$$

We shall show in the following that by means of a similarity transformation it is possible to obtain the solution of equation (17) for the boundary conditions (19).

* This conclusion was drawn on basis of equations valid for small values of the Reynolds number. It remains for the more exact calculation to confirm this conclusion.

Equation (17) and the boundary conditions (19) are compatible with a solution of the form

$$\frac{c}{c_0} = f(\eta), \qquad \eta \equiv \frac{y_1}{\delta}, \qquad \delta = \delta(x_1, z). \tag{20}$$

Indeed, the similarity variable η enables to transform equation (17) into

$$-\left[-\left(\frac{2}{3}\chi - a_0\right)\delta\frac{\partial \delta}{\partial z} + a_0\delta\frac{\partial \delta}{\partial x_1} + \delta^2\frac{\mathrm{d}a_0}{\mathrm{d}z}\right] \times \eta\frac{\mathrm{d}c}{\mathrm{d}\eta} = \frac{2}{3}\frac{Dh_0}{Qk}\frac{\mathrm{d}^2 c}{\mathrm{d}\eta^2}. \tag{21}$$

In order that $c/c_0 = f(\eta)$ and $\delta = \delta(x_1, z)$, one must have

$$-\left(\frac{2}{3}\chi - a_0\right)\frac{1}{2}\frac{\partial \delta^2}{\partial z} + \frac{a_0}{2}\frac{\partial \delta^2}{\partial x_1} + \delta^2\frac{\mathrm{d}a_0}{\mathrm{d}z} = \frac{4}{3}\frac{Dh_0}{Qk}\varepsilon^2 \tag{22}$$

and

$$\frac{\mathrm{d}^2 c}{\mathrm{d}\eta^2} + 2\varepsilon^2\eta\frac{\mathrm{d}c}{\mathrm{d}\eta} = 0 \tag{23}$$

where ε is an arbitrary numerical constant.

Equation (23) and the boundary conditions lead to

$$\frac{c}{c_0} = 1 - \frac{2}{\sqrt{\pi}}\int_0^{\varepsilon\eta} \mathrm{e}^{-\xi^2}\,\mathrm{d}\xi \equiv \operatorname{erfc}\frac{ky_1}{\sqrt{\Delta}} \tag{24}$$

where

$$\Delta \equiv \left(\frac{\delta k}{\varepsilon}\right)^2. \tag{25}$$

The general solution of equation (22) is

$$\frac{3Q}{8kh_0}\frac{\Delta}{D}\left(\frac{2}{3}\chi - a_0\right)^2 + \int_0^z \left(\frac{2\chi}{3} - a_0\right)\mathrm{d}z = \phi(\theta) \tag{26}$$

where ϕ is an arbitrary function of the argument

$$\theta(x_1, z) \equiv x_1 + \int_0^z \frac{a_0\,\mathrm{d}z}{\frac{2}{3}\chi - a_0} \equiv x_1 + I_2(z). \tag{27}$$

The form of the function ϕ may be determined by taking into account that

$$\delta = 0 \quad \text{for} \quad x_1 = 0. \tag{28}$$

Condition (28) is a consequence of the boundary condition (19b) since only if $\delta = 0$ for $x = 0$ equation (24) satisfy the boundary condition (19b).

Using condition (28), equation (26) leads to

$$\phi\left(\int_0^z \frac{a_0\,dz}{\frac{2}{3}\chi - a_0}\right) = \int_0^z \left(\frac{2\chi}{3} - a_0\right) dz \equiv I_1(z). \tag{29}$$

The curve ϕ vs. θ can be traced graphically by observing that to a certain value of the variable z there corresponds a value of ϕ given by equation (29) and a value of θ given by equation (27) (in which x_1 is taken as zero). This curve once traced for several values of the parameter ψ, the quantity Δ and therefore the distribution of concentration are completely determined.

The mass flux at the free interface is given by the equation:

$$N = -D\left(\frac{\partial c}{\partial y_1}\right)_{y_1=0} = \frac{2kDc_0}{\sqrt{(\pi\Delta)}}. \tag{30}$$

Since the mass flux in the case of steady laminar motion is given, for small depths of penetration, by

$$N_l = c_0\sqrt{(3DQ/2\pi H_0 x)}, \tag{31}$$

the amplification factor α, defined by the ratio

$$\alpha = N/N_l, \tag{32}$$

results from

$$\alpha = \left(\frac{8DkH_0x_1}{3Q\Delta}\right)^{\frac{1}{2}}. \tag{33}$$

Taking into account equations (26–28), equation (33) can be written in the form

$$\alpha = \frac{|\frac{2}{3}\chi - a_0|\sqrt{(x_1)}}{\sqrt{[\phi(\theta) - I_1(z)]}}\sqrt{\left(\frac{H_0}{h_0}\right)} = \frac{|\frac{2}{3}\chi - a_0|\sqrt{(x_1)}\,.\sqrt{(H_0/h_0)}}{\sqrt{\{\phi[x_1 + I_2(z)] - \phi[I_2(z)]\}}}. \tag{34}$$

The temporal average of the amplification factor is given by the expression

$$\bar{\alpha} = \frac{1}{2\pi}\left[\sqrt{\left(\frac{x_1 H_0}{h_0}\right)}\right]\int_0^{2\pi} \frac{|\frac{2}{3}\chi - a_0|\,dz}{\sqrt{\{\phi[x_1 + I_2(z)] - \phi[I_2(z)]\}}}. \tag{35}$$

As it is shown in the Appendix the assymptotic value of $\bar{\alpha}$ for sufficiently large values of x (practically $x > 3$ cm) may be calculated by means of the equation

$$\bar{\alpha} = \frac{1}{2\pi}\sqrt{\left[\frac{H_0}{h_0} I_1(2\pi)\,.\,I_2(2\pi)\right]}. \tag{36}$$

For $I_1(2\pi)$ one obtains easily that

$$I_1(2\pi) = 2\pi\left(\frac{2}{3}\chi - 1 + \frac{a^2}{4}A_{02}\right)$$

the values of $I_2(2\pi)$ can be obtained, however, only by numerical integration. Since H_0/h_0, $I_1(\pi)$ and $I_2(2\pi)$ depend only on ψ, the average amplification factor is a function only of ψ. The calculated curve is presented in Fig. 4.

The calculations lead therefore to the conclusion that the average amplification factor depends in the range of validity of the thin layer approximation (low Reynolds numbers) only upon the parameter ψ.

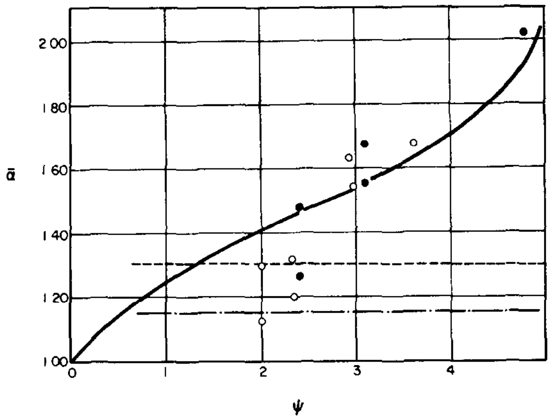

FIG. 4. The average amplification factor α vs. ψ.
----- Author's theoretical curve with Kapitza's velocity distribution
——— Author's theoretical curve with author's velocity distribution [equation (36)].
–·–·– Levich's theoretical curve
○, ● Experimental data of Kamei and Oishi

COMPARISON WITH EXPERIMENT

From their experimental results concerning the rate of mass transfer in a falling liquid film Kamei and Oishi concluded that there exists four regions. A first region $Re \lesssim 35$ for which probably the rate may be predicted by means of the equation valid for the laminar steady flow. A second region $35 < Re < 150$ for which the rate may be predicted if one replaces in the equation valid for the steady laminar case, the diffusion coefficient D by an apparent diffusion coefficient D_a independent of x.

A third region $150 < Re < 1000$ for which no correlation was possible. The fourth corresponds to $Re > 1000$ (turbulent regime).

We notice that the first two regions are in the range of validity of the thin film approximation. It results from our theoretical considerations too that in this range it is possible to use the equation valid for laminar steady motion by replacing the diffusion coefficient by an apparent one, independent on x but depending, *inter alia*, on Q. Outside the range of validity of the thin layer approximation the complexity of the mechanism of mass transfer grows owing to two causes: (1) the wave motion cannot any longer be characterized by means of a single parameter ψ but by means of two parameters ψ and Re; (2) u may become zero or even negative (this has as a consequence the occurrence of circulation motions in the vicinity of the free interface) for a part of the period and it is no longer possible to approximate u in the vicinity of the interface by its value at the interface as in equation (13a). The transition evidenced by Kamei and Oishi in the vicinity of $Re \approx 150$ may therefore be predicted theoretically. Besides the qualitative agreement there exists also a quantitative agreement as results from Fig. 4 where equation (36) is compared with experimental results.

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APPENDIX

For reasons which will become clear in the following let us calculate the average values of α in the points $x_1 = nI_2(2\pi)$ where $n = 1, 2, \ldots$.

Since $a_0(z) = a_0(z + 2\pi)$ one obtains easily that

$$I_1(z + 2\pi) = I_1(z) + I_1(2\pi) \quad \text{(A-1)}$$

and

$$I_2(z + 2\pi) = I_2(z) + I_2(2\pi). \quad \text{(A-2)}$$

Consequently, equation (29) leads to

$$\phi[I_2(z) + I_2(2\pi)] = \phi[I_2(z)] + I_1(2\pi) \quad \text{(A-3)}$$

and by iteration to

$$\phi[I_2(z) + nI_2(2\pi)] = \phi[I_2(z)] + nI_1(2\pi). \quad \text{(A-4)}$$

In the points $x_1 = nI_2(2\pi)$ equation (35) becomes

$$\bar{\alpha}_n = \frac{1}{2\pi}\left\{\sqrt{\left[\frac{H_0 I_2(2\pi)}{h_0 I_1(2\pi)}\right]}\right\}\int_0^{2\pi}\left|\frac{2}{3}\chi - a_0\right|\mathrm{d}z$$

$$= \frac{1}{2\pi}\sqrt{\left[\frac{H_0}{h_0} I_1(2\pi)\, I_2(2\pi)\right]} \quad \text{(A-5)}$$

which is independent upon n.

Let us now take $x_1 \neq nI_2(2\pi)$. It is, however, always possible to write

$$x_1 = nI_2(2\pi) + x_1' \quad \text{where} \quad x_1' < I_2(2\pi). \quad \text{(A-6)}$$

For this point one may write

$$\phi[I_2(z) + x_1' + nI_2(2\pi)] - \phi[I_2(z)] = nI_1(2\pi) + \phi[I_2(z) + x_1'] - \phi[I_2(z)].$$

Since $x_1' < I_2(2\pi)$ and since ϕ is an increasing function on θ,

$$d = \phi[I_2(z) + x_1'] - \phi[I_2(z)] < I_1(2\pi). \quad \text{(A-7)}$$

Consequently

$$\bar{\alpha} = \frac{1}{2\pi}\sqrt{\left[\frac{nI_2(2\pi) + x_1'}{nI_1(2\pi) + d}\cdot\frac{H_0}{h_0}\right]}\, I_1(2\pi)$$

which for sufficiently large values of n (practically for $n \geqslant 2$) lead to:

$$\bar{\alpha} = \frac{1}{2\pi}\sqrt{\left[\frac{H_0}{h_0} I_1(2\pi) \,.\, I_2(2\pi)\right]}.$$

Résumé—Un traitement non-linéaire de l'hydrodynamique du mouvement d'ondes est suggéré et ses résultats sont employés pour la solution du problème du transport de masse. On établit une équation de avec convection dans le cas d'une pènètration faible et on la résout exactement au moyen d'une transformation de similitude.

Le résultat principal obtenu est que, pour de faibles nombres de Reynolds, le facteur d'amplification moyen (moyenne temporelle du rapport entre le flux massique dans le mouvement d'ondes et dans le mouvement laminaire permanent) est une fonction, ayant la forme de la Fig. 4, seulement de la quantité sans dimensions: $\psi = g^{1/9}Q^{11/9}/\nu^{7/9}(\sigma/\rho)^{1/3}$.

Zusammenfassung—Es wird eine nichtlineare Behandlung der hydrodynamischen Wellenbewegung vorgeschlagen und die Ergebnisse zur Lösung des Stoffübergangsproblems herangezogen. Für den Fall geringer Eindringung wird eine Gleichung für konvektive Diffusion aufgestellt und mit Hilfe einer Ähnlichkeitstransformation exakt gelöst.

Als Hauptergebnis zeigt sich, dass für kleine Reynolds-Zahlen der durchschnittliche Verstärkungsfaktor $\bar{\alpha}$ (zeitlicher Mittelwert des Verhältnisses von Massenstrom in Wellenbewegung und in stationärer Laminarbewegung) eine Funktion von der in Abbildung 4 angegebenen Form ist und nur von der dimensionslosen Grösse $\psi = g^{1/9}Q^{11/9}/\nu^{7/9}(\sigma/\rho)^{1/3}$ abhängt.

Аннотация—Рассмотрен нелинейный случай гидродинамики волнового движения, результаты которого используются для решения задачи о массообмене. Составлено уравнение конвективной диффузии, справедливое для частного случая проницаемой поверхности, точное решение которого получено путем автомодельного преобразования.

В результате анализа установлено, что для малых чисел Рейнольдса средний коэффициент усиления (среднее по времени отношение массового потока при волнлвом движении к этой величине в стационарном ламинарном течении) является функцией, имеющей вид, представленный на рис. 4, только при условии, что безразмерная величина

$$\psi = \frac{g^{1/6}\,Q^{11/6}}{\nu^{7/6}(\sigma/\rho)^{1/2}}.$$