Mass Transfer to Falling Films:

Part I. Application of the Surface-Stretch Model to Uniform Wave Motion

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A method is developed for predicting rates of gas absorption into laminar rippling films in terms of the surface velocities. The description is an extension of the surface-stretch model of mass transfer *(1)* and is therefore useful for cases of high Peclet number. The description can be used with any of the presently known hydrodynamic models of rippling films and with any future models which may be developed, provided they satisfy two relatively nonrestrictive conditions: (1) the ripples are of a two dimensional nature, being of constant thickness in the direction normal to their direction of propagation, and having no velocity components in this transverse direction; and (2) the ripples propagate at constant celerity and with constant shape. It can be used for both traveling and standing waves and can be extended to describe the effects of high net-mass transfer rates and combined diffusion and chemical reaction.

Gas absorption by a falling liquid film is of major importance in chemical engineering at the present time, occurring in distillation, gas absorption, humidification, and vapor condensation. However, the complexity of flow patterns in most process equipment makes detailed analysis of the absorption process impossible. As a result, many experimental studies have been made in equipment of simpler geometry, in particular, wetted wall columns.

Even here, however, a complete understanding of the absorption process is known only for the case of laminar, nonrippling flow in the film. When rippling or turbulence is present, available theoretical methods are unable to give even semiquantitative prediction of mass transfer behavior.

This failure of theoretical predictions is due both to insufficient knowledge of fluid-dynamic behavior and to lack of sufficiently powerful methods for integrating the continuity equation.

The only currently available integration method, that of Ruckenstein and Berbente (*13*), fails whenever circulation patterns develop in the ripples (*6*) and is not easily generalized to arbitrary wave motions.

The purpose of the authors is to develop prediction methods which are applicable to any observed surface motion, since many rather different types of motion have been reported (*5, 7, 9*). The current paper, describing mass transfer at high film Peclet number for ripples of constant size, shape, and celerity, represents a first step in this direction.

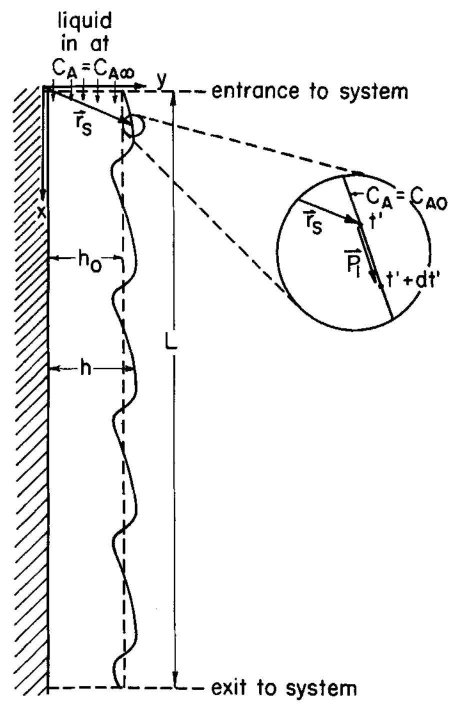

Fig. 1. Physical system.

SYSTEM

A uniform laminar rippling flow of the type pictured in Figure 1 is postulated. By a uniform rippling flow it is meant that each ripple, which may be of an arbitrary shape, propagates at a constant speed or celerity c, with a constant size and shape. That is, the ripple does not speed up or slow down, does not steepen or flatten, and does not grow or shrink in size as it moves down the falling liquid film. The celerity and the velocity profile in the surface region are assumed to be known.

Liquid B enters the system at $x = 0$, with a uniform concentration of dissolved gas A equal to $C_{A\infty}$, but for $x > 0$ the liquid surface is exposed to a gas containing the vapor A. Conditions of operation are such that the concentration of dissolved A at the interface is maintained at C_{AO}, a constant, ($C_{AO} \neq C_{A\infty}$).

Consideration is limited to systems in which the effective depth of penetration of solute, the mass transfer boundary-layer thickness δ_{AB}, is small compared with both the film thickness and the local radius of curvature. These conditions will be met at a sufficiently high film Peclet number $N_{Pe'_1} = h_o v_o / \mathcal{D}_{AB}$. In a system of this type the variation of the tangential component of the liquid velocity over the mass transfer boundary layer will be small, both because of the relatively small shear stresses at the gas-liquid interface and because of the small penetration thickness (*11*).

An adequate description of the mass transfer process is then provided by consideration of each differential surface element separately and the use of the surface stretch modification of the penetration theory (*1*). The equation of continuity for species A and the boundary conditions for a surface element dS then take the form

$$\frac{\partial C_A}{\partial t} + v_n \frac{\partial C_A}{\partial n} = \mathcal{D}_{AB} \frac{\partial^2 C_A}{\partial n^2} \tag{1}$$

$$C_A = C_{A\infty} \quad \text{at} \quad t = t_f \quad \text{for} \quad n > 0 \tag{2}$$

$$C_A = C_{A\infty} \quad \text{at} \quad n \to \infty \quad \text{for} \quad t_f < t < \infty \tag{3}$$

$$C_A = C_{Ao} \quad \text{at} \quad n = 0 \quad \text{for} \quad t_f < t \tag{4}$$

These equations may be integrated in a manner similar to that illustrated in (1) to give

$$dm_A(t') = 2(C_{Ao} - C_{A\infty}) dS_o \sqrt{\mathcal{D}_{AB}/\pi} \times \sqrt{\int_{t_f}^{t_f+t'} [dS(t)/dS_o]^2 \, dt} \tag{5}$$

where $dm_A(t')$ is the total amount of solute transferred across dS in the length of time t', since its entrance into the system:

$$t' = t - t_f \tag{6}$$

The quantity dS_o is a constant reference surface area to be defined presently.

In order to evaluate the rate of mass transfer for the element dS, it is necessary to obtain an explicit expression for the size of the element $dS(t)$ in terms of the fluid velocity profiles and the time spent in the system for each surface element.

ELEMENTAL SURFACE AREA

If the fluid surface element is viewed at a particular instant of time t, then each differential fluid element will have an age t' associated with it. At this instant of time t a surface element can be defined as the rectangle whose four corners coincide with fluid elements associated with t' and $t' + dt'$, and also z and $z + dz$. This is illustrated in Figure 2.

This surface element is always associated with these four imbedded coordinates and will be convected along with them, shrinking and stretching as they move together and apart. The area of particular elements will be a function of the time of observation t and the age of the element t', that is, $dS = dS(t', t)$.

The area of the element whose age is t' at the time of observation t can be obtained as (10)

$$dS(t', t) = |\vec{P}_1 \times \vec{P}_2| \, dt' \, dz \tag{7}$$

where

$$\vec{P}_1 = \left(\frac{\partial \vec{r}_s}{\partial t'}\right)_{z,t} \tag{8}$$

and

$$\vec{P}_2 = \left(\frac{\partial \vec{r}_s}{\partial z}\right)_{t',t} \tag{9}$$

and the position vector $\vec{r}_s$ of the surface is given by

$$\vec{r}_s = x_s \vec{\delta}_x + y_s \vec{\delta}_y + z_s \vec{\delta}_z \tag{10}$$

Assuming a two-dimensional nature for the ripples, so that there is neither dependence on z nor a fluid velocity component in the z direction, one obtains for $\vec{P}_1$ and $\vec{P}_2$:

$$\vec{P}_1 = \left(\frac{\partial x_s}{\partial t'}\right)_{z,t} \vec{\delta}_x + \left(\frac{\partial y_s}{\partial t'}\right)_{z,t} \vec{\delta}_y \tag{11}$$

and

$$\vec{P}_2 = \vec{\delta}_z \tag{12}$$

Therefore, if the x and y coordinates of a given fluid element at the surface are known as a function of its age t' and the time of observation t, then $\vec{P}_1$ and $\vec{P}_2$ could be computed and the area of the surface element $dS(t', t)$ calculated. That is, $x_s(t', t)$ and $y_s(t', t)$ are needed. These must be obtained from the velocity profiles as discussed in the next section.

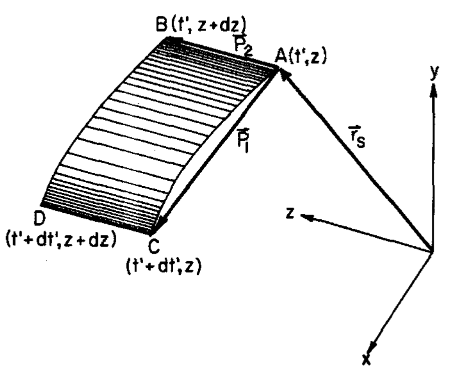

Fig. 2. Surface element.

TRAJECTORIES OF FLUID PARTICLES

For a uniform rippling motion of the type postulated in the first section, the velocity profiles and surface configuration of the film are time independent, as seen by an observer, moving downward at the ripple celerity c. Hence the system velocities and film thickness can be described in terms of the combined variable $\hat{x} = x - ct$ and the variable y:

$$v_x = v_x(\hat{x}, y) \tag{13}$$

$$v_y = v_y(\hat{x}, y) \tag{14}$$

$$h = h(\hat{x}) \tag{15}$$

At the film surface $y = h(\hat{x})$, so that v_{xs} and v_{ys} are functions only of $\hat{x}$. Also, because the surface configuration is time independent to an observer moving along with the ripples, the velocity component normal to the surface, relative to his coordinate system, must be zero. Thus if quantities relative to his ripple-fixed coordinate system are designated by a caret ($\wedge$) so that

$$\hat{x} = x - ct \tag{16}$$

$$\hat{v}_x = \frac{d\hat{x}}{dt} = v_x - c \tag{17}$$

$$\hat{v}_y = v_y \tag{18}$$

then the interrelationship of $\hat{v}_{xs}$ and $\hat{v}_{ys}$ is

$$\frac{\hat{v}_{ys}(\hat{x})}{\hat{v}_{xs}(\hat{x})} = \left(\frac{\partial h}{\partial \hat{x}_s}\right)_z = \left(\frac{\partial y_s}{\partial \hat{x}_s}\right)_z \tag{19}$$

The surface particle trajectories are now obtained by writing

$$\left(\frac{\partial \hat{x}_s}{\partial t}\right)_{z,t_f} = \left[\frac{\partial}{\partial t}(x_s - ct)\right]_{z,t_f} = \left(\frac{\partial x_s}{\partial t}\right)_{z,t_f} - c \tag{20}$$

or

$$\left(\frac{\partial x_s}{\partial t}\right)_{z,t_f} = \hat{v}_{xs}(\hat{x}) - c = \hat{v}_{xs} \tag{21}$$

The subscript s is dropped from this point on, it being understood that all equations refer to conditions at the film surface.

The appropriate boundary condition for integration of Equation (21) is

$$x = 0 \quad \text{at} \quad t = t_f \tag{22}$$

or

$$\hat{x} = -ct_f \quad \text{at} \quad t = t_f \tag{23}$$

Thus

$$\int_{-ct_f}^{\hat{x}} \frac{d\hat{x}}{\hat{v}_x(\hat{x})} = \int_{t_f}^{t} dt = t - t_f \tag{24}$$

This equation gives $x(t, t_f)$ implicitly in terms of the velocity profile which is assumed known. The corresponding relation giving $y(t, t_f)$ could be obtained from Equations (15) and (19). However, as will be shown in the next section, this will not be necessary.

EVALUATION OF SURFACE AREA

The remaining scale vector $\vec{P}_1$ needed to calculate dS can now be determined from Equations (11) and (19):

$$\vec{P}_1 = \left(\frac{\partial x}{\partial t'}\right)_{z,t} \vec{\delta}_x + \left(\frac{\partial y}{\partial t'}\right)_{z,t} \vec{\delta}_y \tag{25}$$

$$\vec{P}_1 = \left(\frac{\partial x}{\partial t'}\right)_{z,t} \left[\vec{\delta}_x + \left(\frac{\partial y}{\partial x}\right)_{z,t} \vec{\delta}_y\right] \tag{26}$$

$$\vec{P}_1 = \left(\frac{\partial x}{\partial t'}\right)_{z,t} \left[\vec{\delta}_x + \left(\frac{\partial y}{\partial \hat{x}}\right)_{z,t} \cdot \left(\frac{\partial \hat{x}}{\partial x}\right)_{z,t} \vec{\delta}_y\right] \tag{27}$$

$$\vec{P}_1 = \left(\frac{\partial x}{\partial t'}\right)_{z,t} \left[\vec{\delta}_x + \frac{\hat{v}_y}{\hat{v}_x} \vec{\delta}_y\right] \tag{28}$$

Equation (24) is now differentiated with respect to t' to obtain

$$\frac{\partial}{\partial t'}(t') = \frac{\partial}{\partial t'}\left[\int_{-ct_f}^{\hat{x}} \frac{d\hat{x}}{\hat{v}_x(\hat{x})}\right] \tag{29}$$

or

$$1 = \frac{1}{\hat{v}_x(\hat{x})}\left(\frac{\partial \hat{x}}{\partial t'}\right)_{z,t} - \frac{1}{\hat{v}_x(-ct_f)} \times (-c) \times \left(\frac{\partial t_f}{\partial t'}\right)_{z,t} \tag{30}$$

But, by virtue of Equations (16) and (6)

$$\left(\frac{\partial \hat{x}}{\partial t'}\right)_{z,t} = \left(\frac{\partial x}{\partial t'}\right)_{z,t} \qquad \left(\frac{\partial t_f}{\partial t'}\right)_{z,t} = -1 \tag{31, 32}$$

Therefore

$$\left(\frac{\partial x}{\partial t'}\right)_{z,t} = \left[1 + \frac{c}{\hat{v}_x(-ct_f)}\right] \hat{v}_x(\hat{x}) \tag{33}$$

Defining

$$\hat{x}_o = -ct_f \tag{34}$$

one has

$$\vec{P}_1 = \left[1 + \frac{c}{\hat{v}_x(\hat{x}_o)}\right] [\hat{v}_x(\hat{x}) \cdot \vec{\delta}_x + v_y(\hat{x}) \cdot \vec{\delta}_y] \tag{35}$$

and then

$$|\vec{P}_1 \times \vec{P}_2| = \left|1 + \frac{c}{\hat{v}_x(\hat{x}_o)}\right| \sqrt{\hat{v}_x^2(\hat{x}) + \hat{v}_y^2(\hat{x})} \tag{36}$$

This expression then permits calculation of the area of an element in terms of its ripple-fixed coordinate $\hat{x}$ and the time it entered the system.

For the reference area dS_o it is convenient to use the flat, nonrippling film, where

$$\hat{v}_x = \frac{3}{2} v_o - c \qquad v_y = 0 \tag{37, 38}$$

Then

$$dS_o = \left|1 + \frac{c}{\frac{3}{2} v_o - c}\right| \times \left|\frac{3}{2} v_o - c\right| dt' dz = \frac{3}{2} v_o \, dt' dz \tag{39}$$

as would be expected. It then follows that

$$\frac{dS}{dS_o} = \frac{2}{3 v_o} \left|1 + \frac{c}{\hat{v}_x(\hat{x}_o)}\right| \sqrt{\hat{v}_x^2 + \hat{v}_y^2} \tag{40}$$

This expression gives the relative stretching and shrinking of the surface element as a function of its ripple-fixed coordinate $\hat{x}_o$ and its entry time t_f. From Equations (16) and (24) $\hat{x}$ will be a function of t and t_f, so that $dS/dS_o = f(t, t_f)$ and we can now evaluate Equation (6). That is

$$dm_A(t') = 2 \cdot \Delta C_A \cdot \frac{3}{2} v_o \, dt' \, dz \sqrt{\frac{\mathcal{D}_{AB}}{\pi}} \times \left|1 + \frac{c}{\hat{v}_x(\hat{x})}\right| \sqrt{\frac{4}{9 v_o^2} \int_{t_f}^{t_f + t'} [\hat{v}_x^2 + \hat{v}_y^2] \, dt} \tag{41}$$

The amount of solute absorbed by an element during its traverse through the system is of most interest. In that case, the age of the element will be the age of an exiting element t_1'.

RATE OF GAS ABSORPTION BY THE SYSTEM

Equation (41) gives the total amount of solute absorbed by an element from the time it enters the system at $t = t_f$ until it leaves the system at $t = t_1$. The rate at which solute is being carried out of the system by the elements is the amount of solute per element times the rate at which elements are leaving the system. That is

$$\frac{dm_A}{dt} = \frac{dm_A}{dS_o} \times \frac{dS_o}{dt} = \frac{dm_A}{dS_o} \times \frac{dS_o}{dt'} \times \frac{dt'}{dt} \tag{42}$$

Therefore, combining Equations (39) and (41) and performing the integration with respect to z, one obtains

$$\left.\frac{dm_A}{dt}\right|_{t=t_1} = 3 W v_o \cdot \Delta C_A \sqrt{\frac{\mathcal{D}_{AB}}{\pi}} \times \left|1 + \frac{c}{\hat{v}_x(\hat{x}_o)}\right| \sqrt{\frac{4}{9 v_o^2} \int_{t_f}^{t_1} [\hat{v}_x^2 + \hat{v}_y^2] \, dt} \tag{43}$$

However, in this form, the equation is difficult to use under most circumstances. It will be convenient to introduce a few substitutions.

First, a change of the variable of integration is made. Thus

$$dt = \frac{dt}{d\hat{x}} \cdot d\hat{x} = \frac{d\hat{x}}{\hat{v}_x(\hat{x})} \tag{44}$$

and the limits of integration becomes

$$\hat{x}(t_1) = x(t_1) - ct_1 = L - ct_1 \equiv \hat{x}_1 \tag{45}$$

and

$$\hat{x}(t_f) = x(t_f) - ct_f = 0 - ct_f \equiv \hat{x}_o \tag{46}$$

Second, it will be convenient to make the factor under the second radical sign dimensionless, through division by the age of an element of a flat film

$$t_o' = 2L/3v_o \tag{47}$$

and multiplication by this factor under the first radical sign.

Third, an instantaneous mass transfer coefficient averaged over the system is defined as

$$k_x = \frac{1}{W \cdot L \cdot \Delta X_A} \cdot \frac{dm_A}{dt} \tag{48}$$

These changes permit Equation (43) to be written as

$$\frac{k_x}{C\, v_o} = \sqrt{\frac{6}{\pi}} \times \sqrt{\frac{\mathcal{D}_{AB}}{L\, v_o}} \times \Lambda \tag{49}$$

where

$$\Lambda = \left| 1 + \frac{c}{\hat{v}_x(\hat{x}_o)} \right| \sqrt{\frac{2}{3\, v_o\, L} \int_{\hat{x}_o}^{\hat{x}_1} \frac{\hat{v}_x^2 + \hat{v}_y^2}{\hat{v}_x}\, d\hat{x}} \tag{50}$$

This equation can now be written in dimensionless form as

$$N_{St_{AB}} = \sqrt{\frac{6}{\pi}} \times N_{Pe'_2}{}^{-1/2} \times \Lambda \tag{51}$$

with $N_{St_{AB}}$ and $N_{Pe'_2}$ being the usual Stanton and length Peclet numbers for mass transfer.

The result of a simple penetration model for a flat, nonrippling film is

$$N_{St_{AB}} = \sqrt{\frac{6}{\pi}}\, N_{Pe'_2}{}^{-1/2} \tag{52}$$

so that the entire effect of the ripples upon the mass transfer can be expressed in the correction factor Λ.

For a given system length, the three quantities L, $\hat{x}_o$, and $\hat{x}_1$ are not all independent. Equations (45) and (46) give

$$t_1 - t_f = \frac{\hat{x}_o + L - \hat{x}_1}{c} \tag{53}$$

but Equation (17) also gives

$$\int_{\hat{x}_o}^{\hat{x}_1} \frac{d\hat{x}}{\hat{v}_x(\hat{x})} = \int_{t_f}^{t_1} dt = t_1 - t_f \tag{54}$$

Therefore

$$\frac{L + \hat{x}_o - \hat{x}_1}{c} = \int_{\hat{x}_o}^{\hat{x}_1} \frac{d\hat{x}}{\hat{v}_x(\hat{x})} \tag{55}$$

This equation then gives the value of $\hat{x}_o$, the initial ripple-fixed coordinate of the surface element, which is determined by a given system length L and final coordinate $\hat{x}_1$.

Equation (55) and (51) then permit calculation of the correction factor Λ for a given value of $\hat{x}_1$. However Λ will generally depend on $\hat{x}_1$, since the rate of mass transfer may depend on whether the element enters or leaves the system on a crest or a trough or somewhere in between. For this reason, the average value of the Stanton number over a long period of exit times is of more interest than the instantaneous values. Thus

$$\overline{N_{St_{AB}}} = \sqrt{\frac{6}{\pi}} \times \overline{N_{Pe'_2}}^{-1/2} \times \overline{\Lambda} \tag{56}$$

where

$$\Lambda = \frac{\int_0^\infty \Lambda\,(t_1)\; dt_1}{\int_0^\infty dt_1} \tag{57}$$

from Equation (45)

$$d\hat{x}_1 = -c\; dt_1 \tag{58}$$

so that

$$\overline{\Lambda} = \frac{\int_0^\infty \Lambda\,(\hat{x}_1)\; d\hat{x}_1}{\int_0^\infty d\hat{x}_1} \tag{59}$$

In general, though, if the ripples have a periodic behavior, then the integrals can be taken over only a single cycle and can be written as

$$\overline{\Lambda} = \frac{\int_0^\lambda \Lambda(\hat{x}_1)\; d\hat{x}_1}{\int_0^\lambda d\hat{x}_1} = \frac{1}{\lambda} \int_0^\lambda \Lambda(\hat{x}_1)\; d\hat{x}_1 \tag{60}$$

In principle, then, the description of gas absorption by the rippling film is complete. Equation (55) permits calculation of $\hat{x}_o$ as a function of $\hat{x}_1$ for a given system length L. Then Equation (50) permits calculation of the instantaneous correction factor as a function of $\hat{x}_1$. Then Equation (59) gives the average correction factor, and finally Equation (51) gives the average Stanton number which describes the average mass transfer coefficient. In actual fact, however, as will be seen in a following section, the equations are extremely complex for any system of real physical interest, and numerical analysis must be employed.

If the rippling motion is periodic, then the basic Equations [(50), (55), and (59)] can be written in dimensionless form by defining the dimensionless quantities

$$\tilde{x} = \hat{x}/\lambda \qquad \tilde{L} = L/\lambda \tag{61, 62}$$

$$\tilde{x}_1 = \hat{x}_1/\lambda \qquad \tilde{x}_o = \hat{x}_o/\lambda \tag{63, 64}$$

$$\tilde{v}_x = \hat{v}_x/v_o \qquad \tilde{v}_y = \hat{v}_y/v_o \qquad \beta = c/v_o \tag{65, 66, 67}$$

Then Equation (55) becomes

$$\frac{\tilde{L} + \tilde{x}_o - \tilde{x}_1}{\beta} \int_{\tilde{x}_o}^{\tilde{x}_1} \frac{d\tilde{x}}{\tilde{v}_x(\tilde{x})} \tag{68}$$

the instantaneous correction factor is

$$\Lambda = \left| 1 + \frac{\beta}{\tilde{v}_x(\tilde{x}_o)} \right| \sqrt{\frac{2}{3\tilde{L}} \int_{\tilde{x}_o}^{\tilde{x}_1} \frac{\tilde{v}_x^2 + \tilde{v}_y^2}{\tilde{v}_x} \, d\tilde{x}} \quad (69)$$

and the average correction factor is

$$\Lambda = \int_0^1 \Lambda(\tilde{x}_1) \, d\tilde{x}_1 \quad (70)$$

These equations describe mass transfer to rippling films [(50), (55), (56), and (59)] and are subject to three restrictions:

1. The ripples are of a two-dimensional nature, being of constant thickness in the z direction.
2. The ripples propagate at constant speed c in the x direction with constant size and shape.
3. The film Peclet number is high enough that $\hat{v}_x$ is essentially constant over the mass transfer boundary layer; this will always occur at sufficiently high Peclet number.

Equations (56), (68), (69), and (70) are further restricted to periodic motion.

Most of the existing hydrodynamic models for rippling films meet the above requirements on flow conditions. The periodic steady state models, [Kapitza (*8*) and Massot, Irani, and Lightfoot (*12*)], the linear stability analyses [Benjamin (*4*) or Yih (*14*)] under conditions of neutral stability, and the nonlinear stability analysis [Anshus (*3*)] all meet these requirements. The equations which have been developed here are not restricted to traveling waves; they are equally applicable to standing waves for which

$$c = 0, \quad \hat{v}_x = v_x, \quad \hat{v}_y = v_y \quad (71, 72, 73)$$

One must, however, be careful in applying these results to long films, where the depth of solute penetration may be large.

EXAMPLE I: FLAT, NONRIPPLING FILM

The first example of the application of the equations will be to a flat, laminar, nonrippling film. Since the penetration model applied to such a film gives Equation (52), should be identically unity everywhere.

For a flat film

$$\tilde{v}_y = 0 \quad (74)$$

$$\tilde{v}_x = (v_x - c)/v_o = \left(\frac{3}{2} v_o - c\right)/v_o = \frac{3}{2} - \beta \quad (75)$$

where β is completely arbitrary. Equation (68) then becomes

$$\frac{\tilde{L} + \tilde{x}_o - \tilde{x}_1}{\beta} = \int_{\tilde{x}_o}^{\tilde{x}_1} \frac{d\tilde{x}}{\frac{3}{2} - \beta} = \frac{\tilde{x}_1 - \tilde{x}_o}{\frac{3}{2} - \beta} \quad (76)$$

This can be rearranged to give

$$\tilde{x}_1 - \tilde{x}_o = \tilde{L} \cdot \left(1 - \frac{2}{3}\beta\right) \quad (77)$$

The correction factor is then given by

$$\Lambda = \left| 1 + \frac{\beta}{\frac{3}{2} - \beta} \right| \sqrt{\frac{2}{3\tilde{L}} \int_{\tilde{x}_o}^{\tilde{x}_1} \left(\frac{3}{2} - \beta\right) d\tilde{x}} \quad (78)$$

or

$$\Lambda = \left| \frac{3/2}{3/2 - \beta} \right| \sqrt{\frac{2}{3\tilde{L}} \cdot \left(\frac{3}{2} - \beta\right) \cdot (\tilde{x}_1 - \tilde{x}_o)} \quad (79)$$

or

$$\Lambda = \left| \frac{1}{1 - \frac{2}{3}\beta} \right| \sqrt{\frac{1}{\tilde{L}}\left(1 - \frac{2}{3}\beta\right) \cdot \tilde{L}\left(1 - \frac{2}{3}\beta\right)} = 1 \quad (80)$$

Thus the equations are consistent in that they give the correct answer for the flat film

EXAMPLE II: PERIODIC STEADY STATE MODEL OF MASSOT, IRANI, AND LIGHTFOOT (MIL), (*12*)

The velocity profile within the film is given by the MIL model as

$$\hat{v}_x = 3v_o \left[\left(\frac{1 + \beta\eta}{1 + \eta}\right) \left(\xi - \frac{\xi^2}{2}\right) - \frac{\beta}{3} \right] \quad (81)$$

and

$$\hat{v}_y - -3\alpha A v_o \sin\left(\frac{2\pi(x - ct)}{\lambda}\right) \left[\left(\frac{1 + \beta\eta}{1 + \eta}\right) \left(\frac{\xi}{2} - \frac{\xi^2}{3}\right) - \left(\frac{\beta - 1}{1 + \eta}\right) \left(\frac{\xi^2}{2} - \frac{\xi^3}{6}\right) \right] \quad (82)$$

where

$$\eta = A \cos\left(\frac{2\pi(x - ct)}{\lambda}\right) \quad (83)$$

is the surface displacement profile and

$$\xi = y/h \quad (84)$$

In addition, the MIL model predicts that α, β, and A are functions only of the Weber number.

If Equations (81) and (82) are evaluated at the surface ($\xi = 1$) and are made dimensionless, they become

$$\tilde{v}_x = \frac{1}{2} \left[\frac{(3 - 2\beta) + \beta\eta}{1 + \eta} \right] \quad (85)$$

$$\tilde{v}_y = \frac{\alpha A}{2} \sin 2\pi\tilde{x} \cdot \left[\frac{(3 - 2\beta) + \beta\eta}{1 + \eta} \right] \quad (86)$$

Equation (68) then becomes

$$\frac{\tilde{L} + \tilde{x}_o - \tilde{x}_1}{\frac{2}{3}\beta} = \int_{\tilde{x}_o}^{\tilde{x}_1} \frac{1 + \eta}{\left(1 - \frac{2}{3}\beta\right) + \frac{1}{3}\beta\eta} \, d\tilde{x} \quad (87)$$

Upon substitution of Equation (83) for η, Equation (87) can be rearranged to give (see Appendix A)

$$\frac{\pi[\tilde{L} + 3(\tilde{x}_o - \tilde{x}_1)]}{\beta - 1} = \int_{2\pi\hat{x}_o}^{2\pi\hat{x}_1} \frac{du}{a + b \cos u} \quad (88)$$

where

$$a = \left(1 - \frac{2}{3}\beta\right), \quad b = \frac{1}{3} A\beta, \quad u = 2\pi\tilde{x} \quad (89, 90, 91)$$

The integration, however, is lengthy and must be carefully performed.

The integral can take several different forms, depending on whether the denominator of the integrand can become zero. This can happen if

$$b^2 \geqq a^2 \tag{92}$$

Examination of Equations (81) and (82) shows that when the denominator of the integrand becomes zero, the surface velocities in the ripple-fixed coordinate system $\hat{v}_x$ and $\hat{v}_y$ also become zero. Equations (81) and (82) can give two types of streamline patterns in the ripple-fixed coordinate system, depending on whether condition (92) is satisfied or not (*6*). These two streamline patterns are shown in Figures 3 and 4.

The circulating type will occur if condition (92) is satisfied, or equivalently if $A > 2 - \frac{3}{\beta}$. The points where $\tilde{v}_x$ and $\tilde{v}_y$ become zero are the stagnation points *A, B, C,* and *C'*. These correspond to the places where the de-

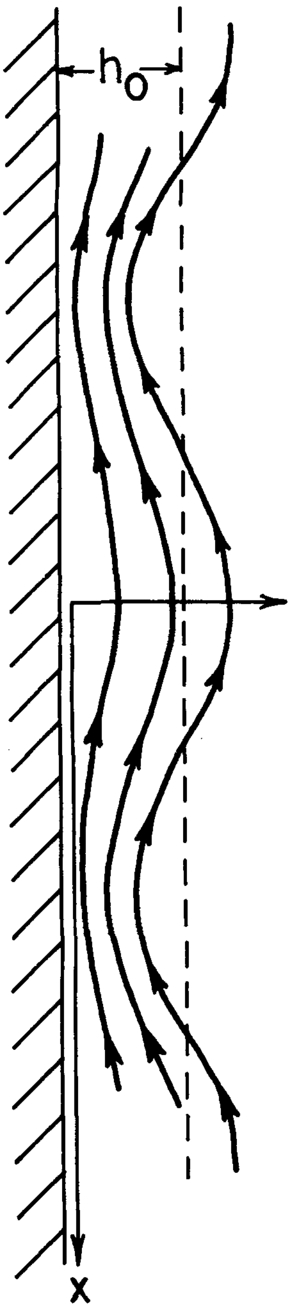

Fig. 3. Noncirculating ripples.

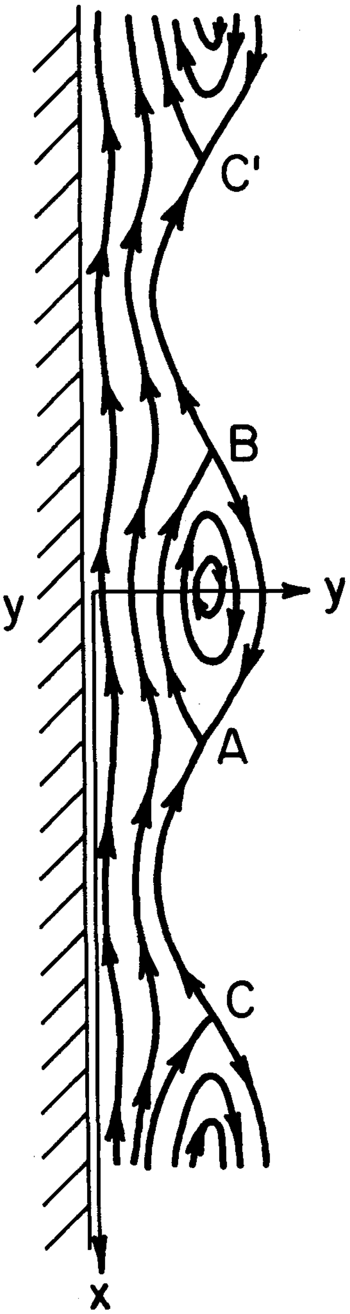

Fig. 4. Circulating ripples.

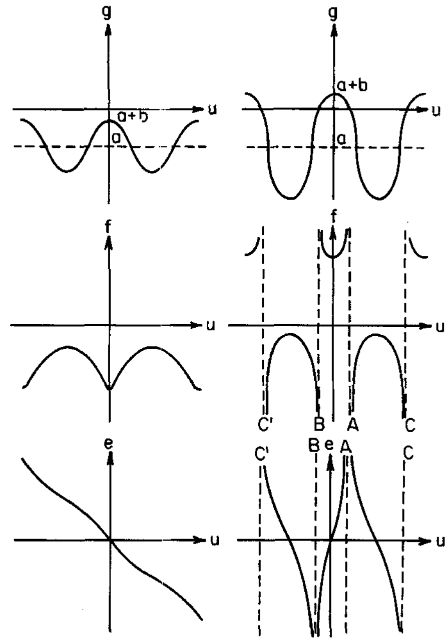

Fig. 5. The functions $g = a + b \cdot \cos(u)$, $f = 1/g$, and $e = \int f\, du$.

nominator of I_1 becomes zero.

The qualitative behavior of the functions $g = a + b \cdot \cos(u)$, $f = 1/g$, and $e = \int f\, du = \int \frac{du}{a + b \cos u}$ are shown in Figure 5 for the two cases $a^2 > b^2$ and $a^2 < b^2$. The points labeled *A, B, C,* and *C'* correspond to similar points in Figure 4.

Because of the physical impossibility of surface elements flowing past the stagnation points, both limits of integration $\tilde{x}_o$ and $\tilde{x}_1$ will lie on the same branch of the *h* curve, so no difficulties will arise from integration across the discontinuities.

Equation (88) can now be written out in closed form for each of the two types of flow (see Appendix A). For the noncirculating flow ($a^2 > b^2$), it is

$$\tan^{-1}\left[\frac{(b-a)\tan \pi\tilde{x}_1}{\sqrt{a^2-b^2}}\right] - \tan^{-1}\left[\frac{(b-a)\tan \pi\tilde{x}_o}{\sqrt{a^2-b^2}}\right] = \frac{\pi[\tilde{L} + 3(\tilde{x}_o - \tilde{x}_1)]\sqrt{a^2-b^2}}{2(\beta-1)} \tag{93}$$

For the circulating flow ($a^2 < b^2$) it is given, in general, by

$$\ln\left|\frac{[(b-a)\tan \pi\tilde{x}_1 + \sqrt{b^2-a^2}]}{[(b-a)\tan \pi\tilde{x}_1 - \sqrt{b^2-a^2}]} \times \frac{[(b-a)\tan \pi\tilde{x}_o - \sqrt{b^2-a^2}]}{[(b-a)\tan \pi\tilde{x}_o + \sqrt{b^2-a^2}]}\right| = \frac{\pi[\tilde{L} + 3(\tilde{x}_o - \tilde{x}_1)]}{\beta-1}\sqrt{b^2-a^2} \tag{94}$$

This last equation can be rewritten in two alternate forms which may be more convenient. In region *AB*

$$\tanh^{-1}\left[\frac{(b-a)\tan \pi\tilde{x}_1}{\sqrt{b^2-a^2}}\right] - \tanh^{-1}\left[\frac{(b-a)\tan \pi\tilde{x}_o}{\sqrt{b^2-a^2}}\right]$$

$$= \frac{\pi[\tilde{L} + 3(\tilde{x}_o - \tilde{x}_1)]\sqrt{b^2 - a^2}}{2(\beta - 1)} \qquad (95)$$

and in regions AC and BC'

$$\coth^{-1}\left[\frac{(b-a)\tan \pi\tilde{x}_1}{\sqrt{b^2 - a^2}}\right] - \coth^{-1}\left[\frac{(b-a)\tan \pi\tilde{x}_o}{\sqrt{b^2 - a^2}}\right]$$
$$= \frac{\pi[\tilde{L} + 3(\tilde{x}_o - \tilde{x}_1)]\sqrt{b^2 - a^2}}{2(\beta - 1)} \qquad (96)$$

These last four equations then give $\tilde{x}_o$ as a function of $\tilde{x}_1$ and $\tilde{L}$ and the hydrodynamic constants, α, β, and Λ, although in an implicit form.

When the velocity profiles of the MIL model, Equations (85) and (86), are substituted into Equation (69), the equation for the instantaneous correction factor Λ, it becomes (see Appendix *B*)

$$\Lambda = \left| \frac{1 + \beta A \cos 2\pi \tilde{x}_o}{\left(1 - \frac{2}{3}\beta\right) + \frac{1}{3} A\beta \cos 2\pi\tilde{x}_o} \right| \sqrt{\frac{1}{2\pi\tilde{L}}} \times \left\{ \frac{2(\beta - 1)[\alpha^2(1 - A^2) - 1]}{\sqrt{1 - A^2}} \left[\tan^{-1}\left(\frac{(1-A)\tan \pi\tilde{x}_1}{\sqrt{1 - A^2}}\right) - \tan^{-1}\left(\frac{(1-A)\tan \pi\tilde{x}_o}{\sqrt{1 - A^2}}\right)\right] \right.$$
$$+ \left(\frac{1}{3}\beta + \alpha^2 + \frac{1}{6}\alpha^2 A^2\beta - \alpha^2\beta\right) \times 2\pi(\tilde{x}_1 - \tilde{x}_o)$$
$$+ \alpha^2 A (\beta - 1)[\sin 2\pi\tilde{x}_1 - \sin 2\pi\tilde{x}_o]$$
$$\left. + \frac{\alpha^2 A^2 \beta}{12}[\sin 4\pi\tilde{x}_1 - \sin 4\pi\tilde{x}_o] \right\}^{1/2} \qquad (97)$$

This equation then gives Λ as a function of $\tilde{x}_o$, $\tilde{x}_1$, $\tilde{L}$ and the hydrodynamic constants of the system α, β, and A. However, $\tilde{x}_o$ is a function of $\tilde{x}_1$, $\tilde{L}$, and the constants, so Λ is a function of $\tilde{x}_1$, $\tilde{L}$, and the constants. If Equation (97) were now averaged over $\tilde{x}_1$, $\overline{\Lambda}$ would be a function only of $\tilde{L}$ and α, β, and A.

However, because of the extreme complexity of Equations (93) through (97), the evaluation of $\overline{\Lambda}$ will necessarily have to be done by numerical analysis. This is being started, and the results will be the subject of a future paper.

From the behavior of the Ruckenstein and Berbente mass transfer model (*13*), it is expected that $\overline{\Lambda}$ will approach a constant value as L becomes large.

HIGH MASS TRANSFER AND CHEMICAL REACTION

Since the surface-stretch model can be extended to predict the effect of high net-mass transfer rate and the effect of simultaneous diffusion and chemical reaction (*2*), and since this extension predicts that these effects are the same as for nonstretching surfaces, it can be stated that these effects will be the same for rippling films as for the flat, nonrippling films, which have been well investigated both theoretically and experimentally.

SUMMARY

Equations (50), (55), (56), and (59) have been developed to predict gas absorption by laminar films which have ripples of constant size and shape, propagating at constant speed in the x direction. The equations can be specialized to Equations (56), (68), (69), and (70) for periodic ripples. These equations have been applied to two examples: the flat film, and the ripple model of Massot, Irani, and Lightfoot (*10*). In the first example, the expected, trivial results ($\Lambda = 1$) are obtained, and in the second example, numerical analysis must be employed for a final description. The effects of high net-mass transfer rate and of chemical reaction are predicted to be the same as for flat, nonrippling films.

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NOTATION

A = amplitude of ripples, dimensionless
a = constant, $a = (1 - 2/3\ \beta)$, dimensionless
b = constant, $b = 1/3\ A\ \beta$, dimensionless
c = ripple celerity, cm./sec.
C = total molar density of solutions, moles/cc.
C_A = concentration of A in liquid, moles/cc.
C_{Ao} = concentration of A in liquid at interface, moles/cc.
$C_{A\infty}$ = concentration of A in liquid far from interface, moles/cc.
$\mathcal{D}_{AB}$ = effective diffusivity of species A through liquid B, sq.cm./sec.
dS = fluid surface element, sq.cm.
dS_o = reference surface element, $dS_o = 3/2\ v_o\ dt'dz$, sq.cm.
e = $\int f\ du$, a function of u, dimensionless
f = $1/g$, a function of u, dimensionless
g = $a + b \cdot \cos u$, a function of u, dimensionless
h = film thickness, cm.
h_o = average film thickness, cm.
I_o = integral defined by Equation (*B*16)
I_1 = integral defined by Equation (*A*12)
I_2 = integral defined by Equation (*B*2)
k_x = instantaneous mass transfer coefficient, moles/sq.cm.-sec.
L = length of system, cm.
$\tilde{L}$ = dimensionless length of the system
m_A = moles of A absorbed by surface element, moles
n = coordinate normal to interface, cm.
$N_{Pe'_1}$ = film Peclet number for mass transfer, $N_{Pe'_1} = h_o v_o/\mathcal{D}_{AB}$
$N_{Pe'_2}$ = length Peclet number for mass transfer, $N_{Pe'_2} = L v_o/\mathcal{D}_{AB}$
$N_{St_{AB}}$ = Stanton number for mass transfer, $N_{St_{AB}} = k_x/C v_o$
$\vec{P}_1$ = displacement vector, defined by Equation (8)
$\vec{P}_2$ = displacement vector, defined by Equation (9)
r = dimensionless constant, $r = \sqrt{b + a/b - a}$
$\vec{r}_s$ = position vector of a point on the liquid surface, cm.
s = dimensionless constant, $s = \sqrt{a + b/a - b}$
t = time, sec.
t' = age of a fluid surface point, sec.

t_f = time at which a given surface element enters the system, sec.
t_1' = age of a surface element when exiting from system, sec.
u = variable of integration, $u = 2\pi\tilde{x}$, dimensionless
v_n = velocity normal to interface, cm./sec.
v_o = average velocity of liquid, cm./sec.
v_x = x component of fluid velocity, relative to fixed coordinates, cm./sec.
$\hat{v}_x$ = x component of velocity, relative to ripple-fixed coordinates, $\hat{v}_x = v_x - c$, cm./sec.
$\tilde{v}_x$ = dimensionless x component of velocity relative to ripple-fixed coordinates, $\tilde{v}_x = \hat{v}_x/v_o$
v_y = y component of fluid velocity, relative to fixed coordinates, cm./sec.
$\hat{v}_y$ = y component of velocity, relative to ripple-fixed coordinates, cm./sec.
$\tilde{v}_y$ = dimensionless y component of velocity relative to ripple-fixed coordinates, $\tilde{v}_y = \hat{v}_y/v_o$
W = width of system, cm.
w = variable of integration, $w = \tan u/2$, dimensionless
x = coordinate down column surface, cm.
$\hat{x}$ = coordinate down column surface, relative to a given ripple, $x = x - ct$, cm.
$\tilde{x}$ = dimensionless coordinate relative to ripple-fixed coordinates, $\tilde{x} = x/\lambda$
$\hat{x}_o$ = initial ripple-fixed x coordinate of a surface element, $\hat{x}_o = -c\, t_f$, cm.
$\hat{x}_1$ = final ripple-fixed x coordinate of a surface element, $\hat{x}_1 = L - ct_1$, cm.
y = coordinate perpendicular to column surface, cm.
z = coordinate across column surface, cm.

Greek Letters

α = dimensionless wave number, $\alpha = 2\pi h_o/\lambda$
β = dimensionless celerity, $\beta = c/v_o$
δ_{AB} = mass transfer boundary-layer thickness, cm.
$\vec{\delta}_x$ = unit vector in x direction
$\vec{\delta}_y$ = unit vector in y direction
$\vec{\delta}_z$ = unit vector in z direction
ΔC_A = total available concentration difference, $\Delta C_A = C_{Ao} - C_{A\infty}$, moles/cc.
ΔX_A = total available mole-fraction difference $\Delta X_A = \Delta C_A/C$, dimensionless
ζ = dummy variable in Equations (A25) and (A26)
η = surface displacement because of ripples, $\eta = A\cos[2\pi(x - ct)/\lambda]$, dimensionless
Λ = instantaneous mass transfer correction factor for effect of ripples, dimensionless
λ = wavelength of periodic ripples, cm.
ξ = local dimensionless coordinate, $\xi = y/h$

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APPENDIX A

Equation (87) is

$$\frac{\tilde{L} + \tilde{x}_o - \tilde{x}_1}{\frac{2}{3}\beta} = \int_{\tilde{x}_o}^{\tilde{x}_1} \frac{1+\eta}{\left(1 - \frac{2}{3}\beta\right) + \frac{1}{3}\eta}\, d\tilde{x} \tag{A1}$$

where

$$\eta = A\cos 2\pi\tilde{x} \tag{A2}$$

Define

$$a = \left(1 - \frac{2}{3}\beta\right) \tag{A3}$$

$$b = \frac{1}{3}A\beta \tag{A4}$$

$$u = 2\pi\tilde{x} \tag{A5}$$

Equation (A1) becomes

$$\frac{\tilde{L} + \tilde{x}_o - \tilde{x}_1}{\frac{2}{3}\beta} = \frac{1}{2\pi}\int_{2\pi\tilde{x}_o}^{2\pi\tilde{x}_1} \frac{1 + A\cos u}{a + b\cos u}\, du \tag{A6}$$

or

$$\tilde{L} + \tilde{x}_o - \tilde{x}_1 = \frac{\beta}{3\pi}\left[\int_{2\pi\tilde{x}_o}^{2\pi\tilde{x}_1} \frac{du}{a + b\cos u} + A\int_{2\pi\tilde{x}_o}^{2\pi\tilde{x}_1} \frac{\cos u\, du}{a + b\cos u}\right] \tag{A7}$$

But

$$\cos u = \frac{(a + b\cos u) - a}{b} \tag{A8}$$

so

$$A\int \frac{\cos u\, du}{a + b\cos u} = \frac{A}{b}\int du - \frac{Aa}{b}\int \frac{du}{a + b\cos u} \tag{A9}$$

Therefore

$$\tilde{L} + \tilde{x}_o - \tilde{x}_1 = \frac{\beta}{3\pi}\left[\frac{A \cdot 2\pi(\tilde{x}_1 - \tilde{x}_o)}{b} + \left(1 - A\frac{a}{b}\right)\int_{2\pi\tilde{x}_o}^{2\pi\tilde{x}_1} \frac{du}{a + b\cos u}\right] \tag{A10}$$

or

$$\tilde{L} + \tilde{x}_o - \tilde{x}_1 = \frac{1}{3\pi}\left[\frac{2\pi}{\frac{1}{3}}(\tilde{x}_1 - \tilde{x}_o) + 3(\beta - 1)\int_{2\pi\tilde{x}_o}^{2\pi\tilde{x}_1} \frac{du}{a + b\cos u}\right] \tag{A11}$$

which can be rearranged to give

$$\frac{\pi[\tilde{L}+3(\tilde{x}_o-\tilde{x}_1)]}{\beta-1}=\int_{2\pi\tilde{x}_o}^{2\pi\tilde{x}_1}\frac{du}{a+b\cos u}=I_1 \qquad (A12)$$

This can now be integrated for two cases, $a^2 > b^2$, and $a^2 < b^2$.

Noncirculating ($a^2 > b^2$)

Let

$$w=\tan u/2 \qquad (A13)$$

$$\therefore \cos u=\frac{1-w^2}{1+w^2} \qquad (A14)$$

$$\therefore du=\frac{2\,dw}{1+w^2} \qquad (A15)$$

Therefore

$$I_1=\frac{2}{a-b}\int_{\tan\pi\tilde{x}_o}^{\tan\pi\tilde{x}_1}\frac{dw}{\left(\frac{a+b}{a-b}\right)+w^2} \qquad (A16)$$

Define

$$s=\sqrt{\frac{a+b}{a-b}} \qquad (A17)$$

Then

$$I_1=\frac{2}{a-b}\int_{\tan\pi\tilde{x}_o}^{\tan\pi\tilde{x}_1}\frac{dw}{s^2+w^2}=\frac{2}{a-b}\times\frac{1}{s}\left[\tan^{-1}\frac{w}{A}\right]_{\tan\pi\tilde{x}_o}^{\tan\pi\tilde{x}_1} \qquad (A18)$$

or

$$I_1=\frac{2}{\sqrt{a^2-b^2}}\left[\tan^{-1}\left(w\sqrt{\frac{a-b}{a+b}}\right)\right]_{\tan\tilde{x}_o}^{\tan\pi\tilde{x}_1} \qquad (A19)$$

or

$$I_1=\frac{2}{\sqrt{a^2-b^2}}\left[\tan^{-1}\left(\frac{|a-b|w}{\sqrt{a^2-b^2}}\right)\right]_{\tan\pi\tilde{x}_o}^{\tan\pi\tilde{x}_1} \qquad (A20)$$

This then yields Equation (93).

Circulating ($a^2 < b^2$)

Again, Equations (A13), (A14), and (A15) are substituted into (A12), but the result is factored differently to give

$$I_1=\frac{2}{b-a}\int_{\tan\pi\tilde{x}_o}^{\tan\pi\tilde{x}_1}\frac{dw}{\left(\frac{a+b}{b-a}\right)-w^2}=\frac{2}{b-a}\int_{\tan\pi\tilde{x}_o}^{\tan\pi\tilde{x}_1}\frac{dw}{r^2-w^2} \qquad (A21)$$

where

$$r=\sqrt{\frac{b+a}{b-a}} \qquad (A22)$$

This integrates to

$$I_1=\frac{2}{b-a}\times\frac{1}{2\pi}\left[\ln\left|\frac{r+w}{r-w}\right|\right]_{\tan\pi\tilde{x}_o}^{\tan\pi\tilde{x}_1} \qquad (A23)$$

or

$$I_1=\frac{1}{\sqrt{b^2-a^2}}\left[\ln\left|\frac{(b-a)w+\sqrt{b^2-a^2}}{(b-a)w-\sqrt{b^2-a^2}}\right|\right]_{\tan\pi\tilde{x}_o}^{\tan\pi\tilde{x}_1} \qquad (A24)$$

This gives Equation (94).

Equations (95) and (96) can be derived from Equation (A24) by use of the two identities

$$\tanh^{-1}=\frac{1}{2}\ln\frac{1+\xi}{1-\xi},\ \xi^2<1 \qquad (A25)$$

and

$$\coth\xi=\frac{1}{2}\ln\frac{\xi+1}{\xi-1},\ \xi^2>1 \qquad (A26)$$

whose applications are straightforward.

APPENDIX B

Upon substitution of Equations (85) and (86) into Equation (69), the result is

$$\Lambda=\left|1+\frac{2\beta[1+\eta(\tilde{x}_o)]}{(3-2\beta)+\beta\eta(\tilde{x}_o)}\right|\times\sqrt{\frac{I_2}{2\pi\tilde{L}}} \qquad (B1)$$

where

$$I_2=2\pi\int_{x_o}^{\tilde{x}_1}\frac{[(3-2\beta)+\beta\eta][1+\alpha^2A^2\sin^2 2\pi\tilde{x}]d\tilde{x}}{3(1+\eta)} \qquad (B2)$$

and

$$\eta(\tilde{x})=A\cos 2\pi\tilde{x} \qquad (B3)$$

Thus

$$\Lambda=\left|\frac{1+A\beta\cos 2\pi\tilde{x}_o}{\left(1-\frac{2}{3}\beta\right)+\frac{1}{3}\beta A\cos 2\pi\tilde{x}_o}\right|\sqrt{\frac{I_2}{2\pi\tilde{L}}} \qquad (B4)$$

where

$$I_2=2\pi\int_{\tilde{x}_o}^{\tilde{x}_1}\frac{\left[\left(1-\frac{2}{3}\beta\right)+\frac{1}{3}A\beta\cos 2\pi\tilde{x}\right][1+\alpha^2\sin^2 2\pi\tilde{x}]d\tilde{x}}{1+A\cos 2\pi\tilde{x}} \qquad (B5)$$

But

$$\sin^2 2\pi\tilde{x}=1-\cos^2 2\pi\tilde{x} \qquad (B6)$$

so

$$I_2\,2\pi\int_{\tilde{x}_o}^{\tilde{x}_1}\frac{\left[\left(1-\frac{2}{3}\beta\right)+\frac{1}{3}A\beta\cos 2\pi\tilde{x}\right][(1-\alpha^2A^2)-\alpha^2A^2\sin^2 2\pi\tilde{x}]d\tilde{x}}{1+A\cos 2\pi\tilde{x}} \qquad (B7)$$

Using substitutions (A3), (A4), and (A5) from Appendix A, we get

$$I_2=2\pi(1+\alpha^2A^2)\int_{2\pi\tilde{x}_o}^{2\pi\tilde{x}_1}\frac{du}{1+A\cos u}+b(1+\alpha^2A^2)\int_{2\pi x_o}^{2\pi\tilde{x}_1}\frac{\cos u\,du}{1+A\cos u}-\alpha^2A^2a\int_{2\pi\tilde{x}_o}^{2\pi\tilde{x}_1}\frac{\cos^3 u\,du}{1+A\cos u}$$

$$- \alpha^2 A^2 b \int_{2\pi\tilde{x}_0}^{2\pi\tilde{x}_1} \frac{\cos^3 u \, du}{1 + A \cos u} \quad (B8)$$

But

$$\cos u = \frac{(1 + A \cos u) - 1}{A} \quad (B9)$$

then

$$\frac{\cos u}{1 + A \cos u} = \frac{1}{A} - \frac{1}{A(1 + A \cos u)} \quad (B10)$$

$$\frac{\cos^2 u}{1 + A \cos u} = \frac{1 + A \cos u}{A^2} - \frac{2}{A^2} + \frac{1}{A^2(1 + A \cos u)} \quad (B11)$$

$$\frac{\cos^3 u}{1 + A \cos u} = \frac{(1 + A \cos u)^2}{A^3} - \frac{3(1 + A \cos u)}{A^3} + \frac{3}{A^3} - \frac{1}{1 + A \cos u} \quad (B12)$$

Therefore

$$\int \frac{\cos u \, du}{1 + A \cos u} = \frac{u}{A} - \frac{1}{A} \int \frac{du}{1 + A \cos u} \quad (B13)$$

$$\int \frac{\cos^2 u}{1 + A \cos u} = \frac{\sin u}{A} - \frac{u}{A^2} + \frac{1}{A^2} \int \frac{du}{1 + A \cos u} \quad (B14)$$

$$\int \frac{\cos^3 u}{1 + A \cos u} = u \left(\frac{1}{A^3} - \frac{1}{2A} \right) - \frac{\sin u}{A^2} + \frac{\sin 2u}{4A} - \frac{1}{A^3} \int \frac{du}{1 + A \cos u} \quad (B15)$$

Define

$$I_o = \int \frac{du}{1 + A \cos u} \quad (B16)$$

Then

$$I_2 = \Big[I_o(\beta - 1)[\alpha^2(1 - A^2) - 1] + \left(\frac{1}{3}\beta + \alpha^2 + \frac{1}{6}\alpha^2 A^2 \beta - \alpha^2 \beta \right) u + \alpha^2 A(\beta - 1) \sin u + \frac{\alpha^2 A^2 \beta}{12} \sin 2u \Big]_{2\pi\tilde{x}_0}^{2\pi\tilde{x}_1} \quad (B17)$$

Since $A < 1$, the integral I_o can be obtained in closed form simply by replacing a by 1 and b by A in Equation ($A19$). This gives

$$I_o = \frac{2}{\sqrt{1 - A^2}} \tan^{-1} \left(\frac{(1 - A) \tan \pi\tilde{x}}{\sqrt{1 - A^2}} \right) \quad (B18)$$

Equations ($B3$), ($B17$), and ($B18$) then yield Equation (97).

Optimal Control of a Distillation Column

C. B. BROSILOW and K. R. HANDLEY

Optimal feedback control has been implemented on a fifteen tray pilot scale rectifying column. The results show that excellent control is obtained in spite of major upsets in the feed flow rate and for large changes in the controller set point. Design and implementation costs for the optimal control system should be competitive with those for standard control systems.

This paper is the first in a projected series of papers on the optimal control of countercurrent separation processes. Binary distillation has been chosen as the first type of countercurrent process to be studied because it presents the fewest difficulties in the design and implementation of the control system. For example, in a binary system, the dynamic state of the process can be estimated by measuring the temperature on every tray. This is an easy operation in comparison to measuring compositions which are required in an absorber or extractor. To make matters easier, this study deals only with the control of the overhead composition in a rectifying section of a column. Work is presently going on to extend results to a full column, and no special problems are anticipated.

Normally the control objective in distillation is to keep the process at the desired steady state in spite of dis-