the 3.94 obtained for the ethyl acetate-water system, considering the accuracy of the measurements, that rounding them all off to 3.9 seems most reasonable:

$$k_C = k_s + 3.9\ (D_A/\theta_C)^{0.5} \qquad (3)$$

Figure 9 shows the comparison of the new data as well as the earlier data *(11)* from baffled vessels with those computed from Equation (3). It is not now possible to recover the information needed to include the earlier data from unbaffled vessels, unfortunately. Most of the data are represented by Equation (3) within 20%. Considering the great number of measurements represented by each plotted point, the scatter is not considered excessive, and the applicability of the proposed mechanism seems reasonably well confirmed. More reliable coalescence data are obviously urgently needed.

ACKNOWLEDGMENT

This work was done under Grant G-21526 of the National Science Foundation, whose support is most gratefully acknowledged.

NOTATION

a = specific interfacial area, sq.ft./cu.ft.
d_p = drop diameter, ft.
D_A = diffusivity of the solute, sq.ft./hr.
I_0/I = incident light intensity/emergent light intensity
k_C = time-average continuous-phase mass transfer coefficient, lb.-moles/hr. (sq.ft.) (lb.-moles/cu.ft.)
k_s = steady state continuous-phase mass transfer coefficient, lb.-moles/hr. (sq.ft.) (lb.-moles/cu.ft.)
N_{Re} = drop Reynolds number
N_{Sc} = continuous-phase Schmidt number
t_C = circulation time, hr.
θ_C = time between coalescences, hr.
μ_C = continuous phase viscosity, lb./(ft.) (hr.)
μ_D = dispersed phase viscosity, lb./(ft.) (hr.)
φ_D = volume fraction dispersed phase

Subscripts

av = for the vessel as a whole
B = bottom of the vessel
c = circulating sphere
M = middle of the vessel
obs = observed
r = rigid sphere
T = top of the vessel

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Dispersion in Laminar Flowing Liquid Films Involving Heat Transfer and Interfacial Shear

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Chemical reactors utilizing downward flowing liquid films have certain advantages when the reaction temperature must be carefully controlled *(1)*. When the reaction is highly exothermic, the wall is cooled and the film is kept thin. Generally, the gas-phase reactant is diluted, and the flows are concurrent to avoid flooding. Chien *(2)* has correlated the static pressure gradient and superficial friction factor with the superficial gas and liquid Reynolds numbers. Stainthorp and Batt *(21)* have investigated the wave properties in a downward concurrent two-phase system. Although many investigations have been performed with falling films (for example see references *4* to *6*), much less

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is known about the flow within thin liquid films which are influenced by shear at the gas-liquid interface and by heat transfer.

In order to predict the behavior of a chemical reactor, a residence time distribution function must be known (*7*, *8*). Tracer studies are often used to determine the validity of a fluid mechanical model. For reasons of economy these studies usually involve the rapid injection of a small amount of tracer at the reactor entrance, and monitoring the fluid at the reactor exit. Since the F diagram, the internal age distribution function, and the exit age distribution function are interrelated in a simple manner, knowledge of any one function allows the determination of the other two functions. As pointed out by Danckwerts (*7*), the F diagram (or F function) is easy to obtain, and permits certain calculations to be made concerning the performance of the system when used, for instance, as a blender or reactor.

Consequently, the solution to the problem under consideration will be presented in terms of the F diagram or F function. It is noted that the F function is simply that integral of the exit age distribution function E.

G. I. Taylor (*9*) established the analytical foundation concerning the treatment of dispersion in which both convection and diffusion are important. Although several significant extensions have been made to the theory of such dispersions (*10* to *13*), the laminar Newtonian flow in a film when influenced by both heat transfer and interfacial shear has not been available previously.

After the general problem is stated, two limiting cases are solved. These two limiting cases appear when $L/U_1 >> \Delta^2/\pi^2 D_0$ and when $L/U_1 << \Delta^2/\pi^2 D_0$ where L is the length of the reactor, U_1 is the velocity at the gas-liquid interface, Δ is the average film thickness, and D_0 is the minimum value of the molecular diffusion coefficient of the tracer which is introduced into the liquid stream at the top of the reactor. In order to account for the influence of the concurrent flow of gas, both limiting cases are solved for an arbitrary shear stress at the gas-liquid interface. In practice, the shear stress at the gas-liquid interface must be determined from the measured pressure drop through the reactor. A momentum balance yields $\tau_1 = R\Delta P/2L$ where R is the distance from the tube center to the gas-liquid interface and ΔP is the measured pressure drop over the length L. Nicklin and Koch (*14*) have proposed a theoretical model for countercurrent systems which relates the film thickness to the interfacial shear stress; it appears that this model may be extended to concurrent systems. Until either the Nicklin-Koch model is extended or other models proposed, the film thickness in concurrent systems must be determined either from experiment or from empirical correlation.

The purpose of this work is to predict the dispersion of a tracer in a laminar flowing film for which the interfacial shear stress and the film thickness have been determined a priori. The solution provides a theoretical basis upon which experimental information may be used to determine whether or not the liquid flow in a specific system is laminar. Conversely, if the film is known to be in laminar motion, calculated distribution curves can be compared with corresponding experimental data to yield the average film thickness.

The analysis is performed for a small linear temperature drop across the film. The viscosity is allowed to vary exponentially with temperatures. The diffusion coefficient is also allowed to vary in such a manner that the group $D\mu/T$ is a constant. For comparison, the highly stressed film is analyzed for the case in which the viscosity is allowed to vary exponentially with temperature, but in which the diffusion coefficient is constant.

GENERAL STATEMENT OF PROBLEM

Consider the steady state flow in a laminar liquid film as shown in Figure 1. The temperature at any point within the film satisfies the energy equation

$$d^2T/dy^2 = 0 \tag{1}$$

under the boundary conditions

$$T = T_0 \quad \text{at} \quad y = 0 \tag{2}$$

and

$$T = T_1 \quad \text{at} \quad y = \Delta \tag{3}$$

The velocity at any point within the film satisfies the momentum equation

$$\frac{d}{dy}\left(\mu(y)\,\frac{dU}{dy}\right) = -\,\rho\, g \cos\theta \tag{4}$$

under the boundary conditions

$$U = 0 \quad \text{at} \quad y = 0 \tag{5}$$

and

$$\mu\frac{dU}{dy} = \tau_1 \quad \text{at} \quad y = \Delta \tag{6}$$

At time equal to zero, a small amount of liquid tracer is uniformly injected into the film at $x = 0$. The concentration of the tracer satisfies the diffusion equation

$$\frac{\partial c}{\partial t} + U(y)\,\frac{\partial c}{\partial x} = \frac{\partial}{\partial y}\left(D(y)\,\frac{\partial c}{\partial y}\right) \tag{7}$$

under the boundary conditions

$$\partial c/\partial y = 0 \quad \text{at} \quad y = 0 \tag{8}$$

and

$$\partial c/\partial y = 0 \quad \text{at} \quad y = \Delta \tag{9}$$

The initial condition is

$$C = \delta(x) \quad \text{at} \quad t = 0 \quad \text{for} \quad 0 \leq y \leq \Delta \tag{10}$$

where $\delta(x)$ represents the delta function.

As mentioned by Farrell and Leonard (*12*), efficient operation of some flow systems often requires that the holding times be confined to a narrow range. The more nearly the holding time is confined to a narrow range, the more the F diagram appears as a step function. For the problem under consideration, the F diagram is defined as

$$F(t, x = L > 0) = \int_{t=0}^{t} \int_{y=0}^{\Delta} \left(UC - D\frac{\partial c}{\partial y}\right) dydt \tag{11}$$

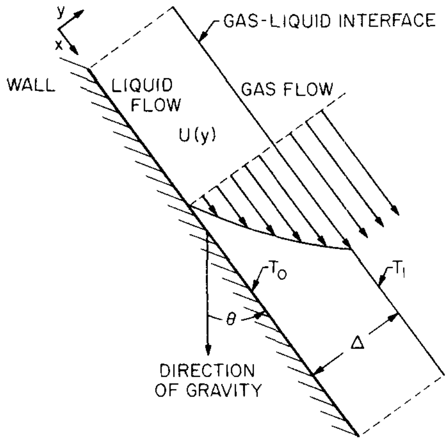

Fig. 1. Diagram of flow system.

This function also represents the fraction of material introduced into the system at $x = 0$ and $t = 0$, which appears at $x = L$ between $t = 0$ and t.

The first product (UC) in Equation (11) represents the dispersion of particles which travel along streamlines with unequal speeds. The product $\left(D\frac{\partial c}{\partial y}\right)$ in Equation (11) represents the effect of ordinary diffusion normal to the streamlines. When the characteristic time associated with convective transport is much shorter than that for diffusive transport, the problem reduces to one of purely convective dispersion (*15*). When the time associated with convective transport is near that associated with diffusive transport, analytical solutions are difficult to obtain. However, many cases of commercial interest involve systems in which the characteristic time associated with convective transport is much longer than that associated with diffusive transport. Consequently, this limiting case will be treated in detail.

Taylor Dispersion

The first and second terms in Equation (7) indicate that the characteristic time associated with convective transport is of order L/U_1. The first and last terms in Equation (7) indicate that the characteristic time associated with diffusive transport is of order $\Delta^2/\pi^2 D_0$. We shall now obtain the solution to the above-mentioned problem when $L/U_1 >> \Delta^2/\pi^2 D_0$.

Solution of Equations (1), (2), and (3) yields

$$T = T_0 + (T_1 - T_0)\eta \qquad (12)$$

Near atmospheric pressure most Newtonian liquids possess a viscosity-temperature dependence of the form

$$\mu = Me^{N/T} \qquad (13)$$

where M and N are constant and positive. When $(T_1 - T_0)/T_0 << 1$, substitution of Equation (12) into Equation (13) and rearrangement yield

$$\mu = \mu_0 e^{-\alpha\eta} \qquad (14)$$

where $\alpha = -N(T_1 - T_0)/T_0^2$. In the molecular description of viscosity, the quantity N represents a free energy of activation (*16, 17*); Trouton's rule leads to $\alpha \sim 3.8$ $T_b(T_1 - T_0)/T_0^2$ where T_b is the boiling point of the hydrocarbon. Typical hydrocarbon systems involve liquids with $T_b/T_0 < 2$. Consequently, for values of $(T_1 - T_0)/T_0 \sim 0.1$, typical values of α will be less than unity. As shown in Figure 2, very high values of α and low values of β give rise to s-shaped velocity profiles within the film. Increasing values of α correspond to increasing variations of viscosity throughout the film. The value of $\alpha = 0$ corresponds to the isothermal case.

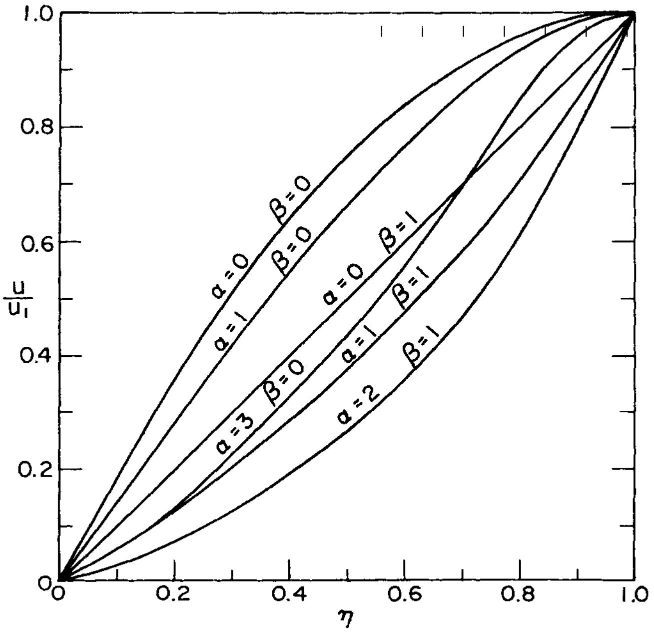

Fig. 2. Velocity profiles in film as a function of α and β.

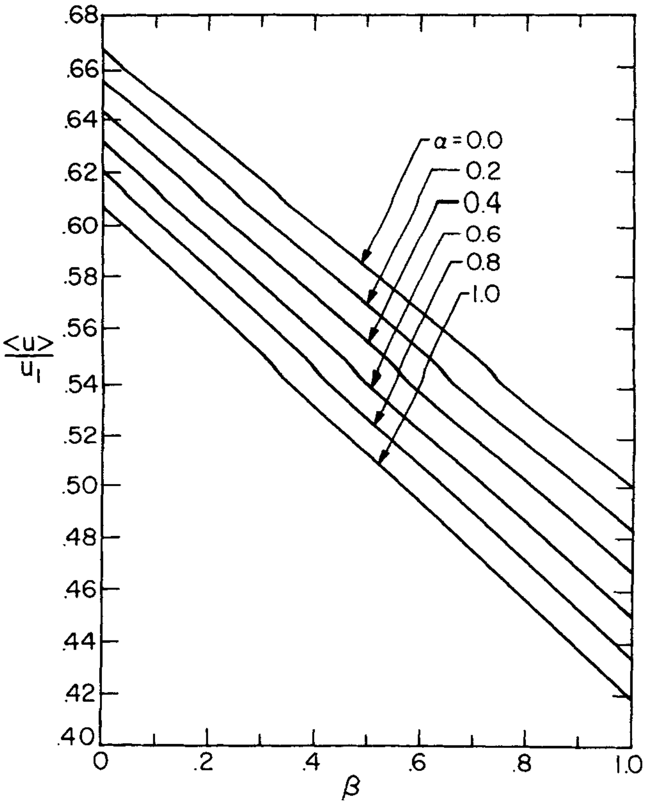

Fig. 3. Average velocity in film as a function of α and β.

Wilke (*18*) has demonstrated that for many liquid binary systems, the group $D\mu/T$ is essentially independent of temperature. Thus, use of Equations (12) and (13) yields

$$D = D_0 e^{\alpha\eta} \qquad (15)$$

when $(T_1 - T_0)/T_0 << 1$.

Unfortunately, very little binary diffusion data exist for highly viscous systems. Although Equation (13) is expected to describe adequately the temperature dependence of viscosity for highly viscous systems, the binary diffusion coefficient may not vary so as to make the group $D\mu/T$ independent of temperature. For comparison, the case of constant diffusion coefficient is reported in Appendix A. When possible, it is recommended that the constancy of the group $D\mu/T$ be checked when applying this analysis to highly viscous systems.

The velocity profile associated with the film is obtained by substituting Equation (14) into Equation (4) and solving the resulting expression under the appropriate boundary conditions. The result is

$$\frac{U}{U_1} = (1 - \beta)\left[\frac{\alpha\eta e^{\alpha\eta} - \alpha e^{\alpha\eta} - e^{\alpha\eta} + \alpha + 1}{1 + \alpha - e^{\alpha}}\right] + \beta\left[\frac{e^{\alpha\eta - 1}}{e^{\alpha} - 1}\right] \qquad (16)$$

where $\beta = \frac{\Delta}{\mu}(\tau_1/U_1)(e^{\alpha} - 1)/\alpha$. The value of $\beta = 0$

corresponds to the falling film case. For large values of the interfacial shear τ_1, the velocity at the interface U_1 tends to the value of $(\tau_1\Delta/\mu)(e^{\alpha}-1)/\alpha$; consequently, the value of $\beta = 1$ corresponds to a highly stressed film. As shown in Figure 2, typical variations in the values of α and β give rise to large variations in the velocity profile. When $\alpha = 0$, Equation (16) reduces to the familiar expression for the isothermal film:

$$\frac{U}{U_1} = (\beta - 1)\eta^2 + (2 - \beta)\eta \tag{17}$$

Values of $\beta = 0$ and 1 correspond to the falling film and the highly stressed film (Couette profile), respectively.

The average velocity, used later in the analysis, is obtained by integrating Equation (16) from $\eta = 0$ to $\eta = 1$. The result is

$$\frac{<U>}{U_1} = (1 - \beta)\left[\frac{\alpha^2 + 2\alpha + 2 - 2e^{\alpha}}{\alpha^2 + \alpha - \alpha e^{\alpha}}\right] + \beta\left[\frac{e^{\alpha} - \alpha - 1}{\alpha e^{\alpha} - \alpha}\right] \tag{18}$$

As shown in Figure 3 the ratio of the average velocity to the maximum velocity may range from 0.42 to 0.66 for typical values of α and β. When $\alpha \to 0$ Equation (18) tends to

$$\frac{<U>}{U_1} = \frac{4 - \beta}{6} \tag{19}$$

We shall now consider a plane which is perpendicular to the velocity field, and which moves with the constant speed $<U>$. Thus, use of the transformation

$$X_* = x - <U>t \tag{20}$$

in Equation (7) yields

$$\frac{\partial}{\partial \eta}\left(D\frac{\partial c}{\partial \eta}\right) = (U - <U>)\left(\frac{\partial c}{\partial X_*}\right)\Delta^2 \tag{21}$$

Substitution of Equations (15), (16), and (18) into Equation (21) and solving the resulting expression under the appropriate boundary condition yields

$$C = C(0) + A(1 - \beta)\left\{\frac{[(2\alpha^2 + 4\alpha - 4\alpha e^{\alpha})\eta - (4e^{\alpha} + 2\alpha^2 + 2\alpha - 4)]e^{-\alpha\eta}}{2\alpha^3(1 + \alpha - e^{\alpha})} + \frac{\alpha^3\eta^2 - (2\alpha^3 + 4\alpha^2)\eta + (4e^{\alpha} + 2\alpha^2 + 2\alpha + 4)}{2\alpha^3(1 + \alpha - e^{\alpha})}\right\}$$
$$+ A\beta\left\{\frac{[\alpha(e^{\alpha} - 1)\eta + (e^{\alpha} + \alpha - 1)]e^{-\alpha\eta} + \alpha^2\eta - (e^{\alpha} + \alpha - 1)}{\alpha^3(e^{\alpha} - 1)}\right\} \tag{22}$$

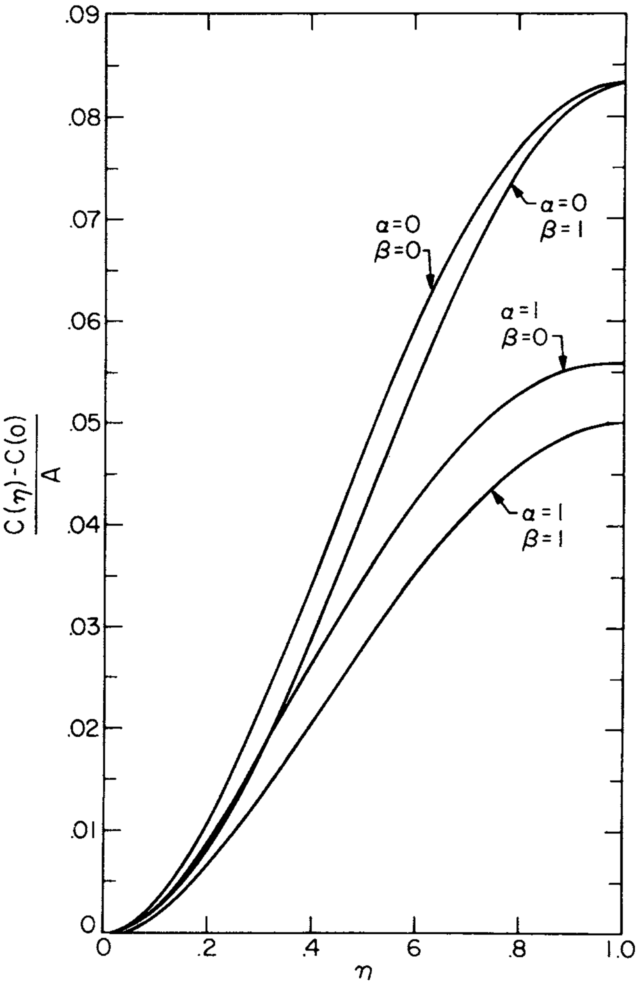

Fig. 4. Tracer concentration profile in film at a fixed X_* for various values of α and β.

where $C(0)$ represents the value of C at $\eta = 0$, and the coefficient $A = (U_1\Delta^2/D_0)\dfrac{\partial C(0)}{\partial X_*}$. The quantity $C(y)$ presented in Equation (22) represents the concentration profile measured in a reference frame moving with the mass average velocity. Typical concentration profiles are shown in Figure 4; as α increases in value, the concentration of tracer becomes more uniform in the direction perpendicular to the flow. When $\alpha \to 0$, a somewhat tedious calculation shows that Equation (22) tends to

$$C = C(0) + \frac{A}{12}[\beta(\eta^4 - 2\eta^3 + \eta^2) - (\eta^4 - 4\eta^3 + 4\eta^2)] \tag{23}$$

As expected Equation (23) is also the result of solving Equations (15), (17), and (19) under the appropriate boundary conditions.

Since we have neglected the longitudinal diffusion, the rate of transfer of the tracer across the plane at X_* is

$$Q_* = \Delta\int_0^1 (U - <U>)\,Cd\eta \tag{24}$$

Since variations of C in the η direction are small compared with those in the X direction

$$\frac{\partial C(0)}{\partial X_*} \simeq \frac{\partial <C>}{\partial X_*} \tag{25}$$

where

$$<C> = \int_0^1 Cd\eta \tag{26}$$

The quantity $<C>$ satisfies the following expression of continuity:

$$\frac{\partial Q_*}{\partial X_*} = -\Delta\frac{\partial <C>}{\partial t} \tag{27}$$

where the differentiation with respect to t is at a point where X_* is constant. Substitution of Equation (24) into Equation (27) yields the equation which governs the longitudinal dispersion of the tracer:

$$\frac{\partial <C>}{\partial t} = k \frac{\partial^2 <C>}{\partial X_*^2} \tag{28}$$

where

$$k = \left(\frac{U_1^2 \Delta^2}{D_0} \right) I(\alpha, \beta) \tag{29}$$

and

$$I(\alpha, \beta) = - \frac{1}{A U_1} \int_0^1 (U - <U>)\, C d\eta \tag{30}$$

The quantities U, $<U>$, and C are given by Equations (16), (18), and (22), respectively.

In Taylor dispersion problems, the integral represented by Equation (30) is usually the bottleneck in obtaining the solution. In this case $I(\alpha, \beta)$ reflects the influence of thermal gradients and interfacial shear upon the dispersion coefficient k. Typical results are shown in Figure 5; by interpolation with Figure 5, values of the dispersion coefficient may be obtained relatively rapidly. Once the dispersion coefficient is known, the residence time distributions may be calculated. A rather tedious calculation yields

$$\begin{aligned} I(\alpha, \beta) = {} & (1-\beta)^2 \\ & \left\{ \left[\frac{48e^{2\alpha} - (4\alpha^4 + 24\alpha^3 - 60\alpha^2 + 48\alpha + 144)e^{\alpha}}{6\alpha^5 (1 + \alpha - e^{\alpha})^2} \right] \right. \\ & \left. + \left[\frac{-(12\alpha^2 + 48\alpha + 48)e^{-\alpha} - (4\alpha^5 + 20\alpha^4 + 48\alpha^3 + 48\alpha^2 - 96\alpha - 144}{6\alpha^5 (1 + \alpha - e^{\alpha})^2} \right] \right\} \\ & - \beta(1 - \beta \left\{ \frac{24e^{2\alpha} - (\alpha^4 + 12\alpha^3 - 12\alpha^2 + 12\alpha + 72)e^{\alpha} - (12\alpha + 24)e^{-\alpha} - (5\alpha^4 + 12\alpha^3 + 12\alpha^2 - 24\alpha - 72)}{3\alpha^5 (1 + \alpha - e^{\alpha})(e^{\alpha} - 1)} \right\} \\ & + \beta^2 \left\{ \frac{2e^{2\alpha} - (\alpha^3 + 6)e^{\alpha} - 2e^{-\alpha} - (\alpha^3 - 6)}{\alpha^5 (e^{\alpha} - 1)^2} \right\} \end{aligned} \tag{31}$$

For α greater than about 0.1, variations of $I(\alpha, \beta)$ with β become significant. The dashed line in Figure 5 represents the solution to the hypothetical case in which $\mu = \mu_0 e^{-\alpha\eta}$ but $D = D_0$ (see Appendix A). Thus, when based upon the maximum velocity, the virtual coefficient of diffusion is influenced primarily by changes in the molecular diffusion coefficient. When $\alpha \rightarrow 0$, another tedious calculation shows that Equation (31) tends to

$$I(0, \beta) = \frac{\beta^2 - 2\beta + 64}{7560} \tag{32}$$

As expected Equation (32) is also the result of solving Equation (30) when the quantities U, $<U>$, and C are given by Equations (17), (19), and (23), respectively. When $\alpha = 0$, the value of I ranges from $I(0, 1) = 1/120$ to $I(0, 0) = 8/945$; thus $I(0, \beta)$ varies less than 1.6% between the highly stressed film and the falling film.

As indicated by Levenspiel and Smith (*18*), the solution to Equation (28) is

$$\frac{<C>V}{Q} = (4\pi k t / L^2)^{-1/2} \, e^{-X_*^2/4kt} \tag{33}$$

The quantity Q represents the volume of tracer of unit concentration which corresponds to the amount of tracer introduced into the system. The quantity V represents the fluid volume in the system located between $x = 0$ and $x = L$. A plot of $<C>V/Q$ versus $<U>t/V$, known as the C diagram, has been discussed elsewhere (for example, see reference 7). As pointed out by Danckwerts (7), the

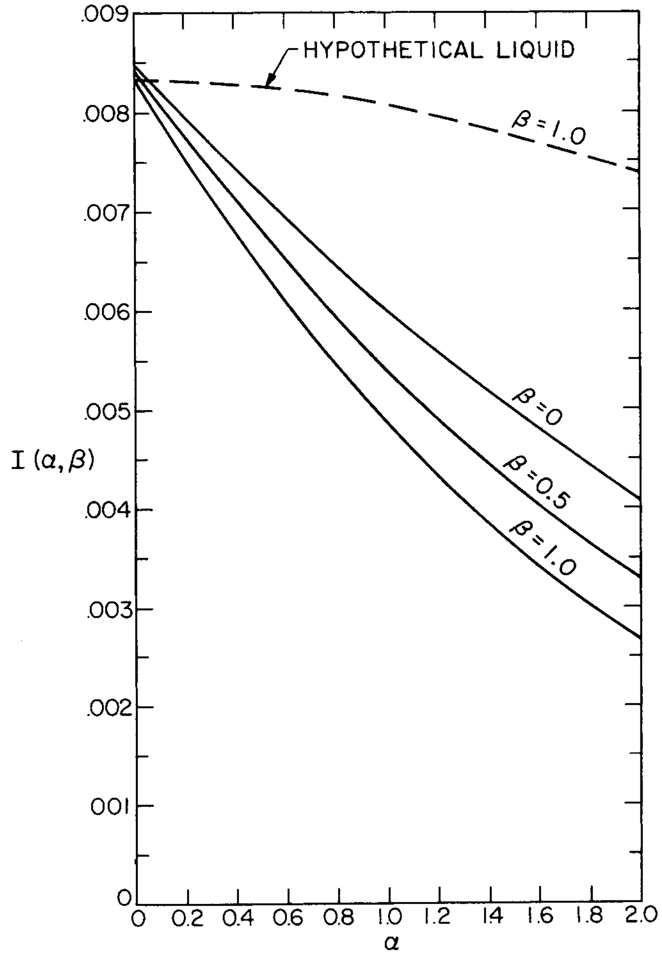

Fig. 5. $I(\alpha, \beta)$ versus α for various β.

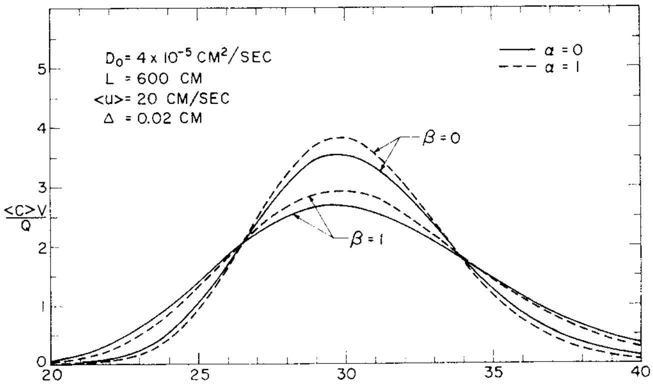

Fig. 6. Plot of $<C>V/Q$ versus t for typical case in which $L/U_1 >> \Delta^2/\pi^2 D_0$.

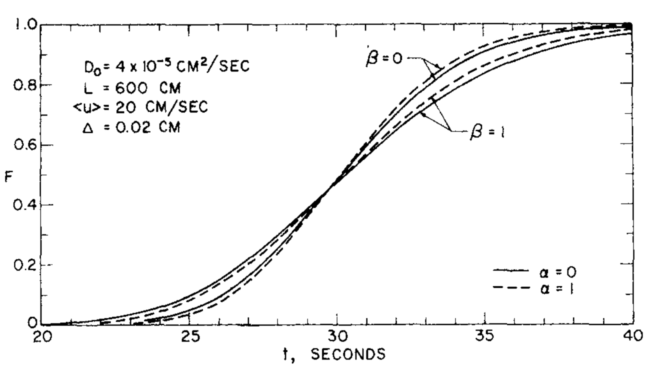

Fig. 7. Plot of F function versus t for typical case in which $L/U_1 >> \Delta^2/\pi^2 D_0$.

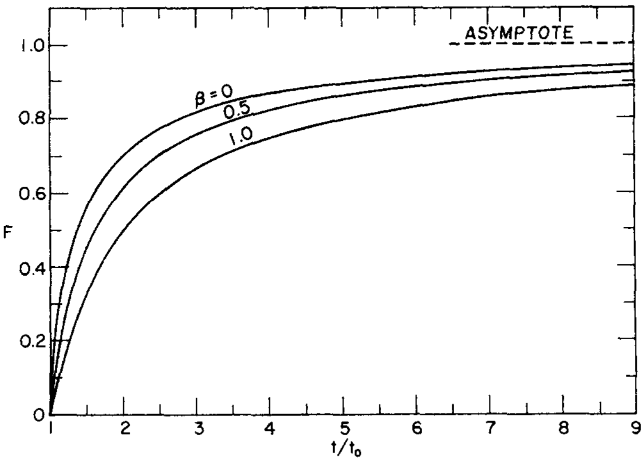

Fig. 8. Plot of F function versus t/t_0 for typical case in which $L/U_1 << \Delta^2/\pi^2 D_0$.

exit age distribution function is equal to $<C><U>/Q$. A plot of $<C>V/Q$ versus t is shown in Figure 6 for a typical system.

Figure 6 indicates that with larger temperature drops through the film, the more the distribution function is driven toward plug flow. Also, the higher the interfacial shear, the more the distribution function is driven away from plug flow. It should also be noted that the interfacial shear exerts a slightly greater influence upon the distribution function than does the temperature drop through the film.

The function $F(t, x = L > 0)$ is obtained by integrating $<C>V/Q$ from $<U>t/L = 0$ to $<U>t/L$ (8). The result is

$$F(t, x = L > 0)$$

$$= \frac{1}{2}\left\{ e^{\frac{<U>L}{k}} \left[\operatorname{erf}\left(\frac{<U>t^{1/2}}{2k^{1/2}} + \frac{Lt^{-1/2}}{2k^{1/2}} \right) - 1 \right] + \operatorname{erf}\left(\frac{<U>t^{1/2}}{2k^{1/2}} - \frac{Lt^{-1/2}}{2k^{1/2}} \right) + 1 \right\} \quad (34)$$

Typical results are shown in Figure 7. As expected the F versus t curves are S-shaped when $L/U_1 >> \Delta^2/\pi D_0$. It is important to note that the point corresponding to the mean residence time of the tracer (that is, $F = 0.5$) is relatively insensitive to heat transfer and interfacial shear (that is, values of α and β), but quite sensitive to the mean residence time of the liquid; furthermore, the mean residence time of the tracer corresponds to the mean residence time of the liquid within a few percent. Consequently, the determination of the average film thickness by the tracer technique is expected to be quite accurate in systems involving heat transfer and interfacial shear. The length of the tube divided by the time corresponding to $F = 0.5$ yields the average velocity of the liquid; the volumetric flow rate per wetted perimeter divided by the average velocity of the liquid yields the average film thickness.

Kim and Knudsen (*20*) investigated a climbing film flow for air and water in an annular duct. They concluded that their system contained a laminar wavy film driven by a turbulent gas flow. Therefore it is reasonable to expect that in concurrent downward flow, the liquid film will be in laminar flow for very low liquid Reynolds numbers even though the gas flow may be turbulent. If the film is known to be in laminar flow, this analysis allows the film thickness to be determined from tracer experiments. Knowledge of the film thickness is especially important in highly exothermic systems. Conversely, if the film thickness is determined a priori, it is expected that this analysis will allow a test to be made in order to determine whether or not the film is in laminar flow.

Convective Dispersion

We shall now obtain the solution to the problem when $L/U_1 << \Delta^2/\pi^2 D_0$ (for details see reference *15*). Since molecular diffusion is neglected, the distance traveled by a particle of fluid tracer is $X = Ut$. Thus the fraction of tracer which appears at a distance $X = L$ downstream between $t = t_0$ and $t \geqq t_0$ is equal to $(1 - \eta_*)$, where η_* is the solution to

$$t_0/t = (1 - \beta) \left[\frac{\alpha\eta_* e^{\alpha\eta_*} - \alpha e^{\alpha\eta_*} + \alpha + 1}{1 + \alpha - e^{\alpha}} \right] + \beta \left[\frac{e^{\alpha\eta_*} - 1}{e^{\alpha} - 1} \right] \quad (35)$$

and

$$t_0 = L/U_1 \quad (36)$$

Since this case appears to be of less commercial interest than the preceding case, calculations will be performed only for the case of $\alpha = 0$. When $\alpha = 0$

$$F(t, X = L > 0) = 0 \quad \text{for} \quad t < t_0 \quad (37)$$

and

$$F(t, X = L > 0)$$

$$= 1 - \left\{ \frac{\left(\frac{2-\beta}{1-\beta}\right) - \left[\left(\frac{2-\beta}{1-\beta}\right)^2 - \frac{4(t_0/t)}{1-\beta} \right]^{1/2}}{2} \right\} \quad \text{for} \quad t \geqq t_0 \quad (38)$$

When $\beta = 0$, Equation (38) reduces to the solution for the falling film:

$$F(t, X = L > 0) = (1 - t_0/t)^{1/2} \quad \text{for} \quad t \geqq t_0 \quad (39)$$

When $\beta = 1$, Equation (38) reduces to the solution for Couette flow:

$$F(t, X = L > 0) = (1 - t_0/t) \quad \text{for} \quad t \geqq t_0 \quad (40)$$

As shown in Figure 8 when $L/U_1 << \Delta^2/\pi^2 D_0$ the F versus t curves are C-shaped. Here again increasing the interfacial shear drives the distribution function away from plug flow. Also, it should be noted that the distribution curves involving Taylor dispersion are much closer to plug flow (based upon the average velocity) than those associated with purely convective dispersion. The distribution function in purely convective dispersion rises more rapidly in the beginning and tails off less rapidly in the end, as compared with that for Taylor dispersion.

CONCLUDING REMARKS

The dispersion of a tracer in a laminar flowing Newtonian liquid film is analyzed for two limiting cases. The limiting cases occur when the characteristic time for convective transport of the tracer is much larger and much smaller than that associated with diffusive transport. The gas-liquid interface is allowed to support a finite shear stress resulting from a concurrent flow of gas. A small linear temperature drop is allowed to exist between the gas-liquid interface and the wall. The viscosity of the liquid is taken to vary exponentially with temperature. The molecular diffusion coefficient is taken to vary in such a manner that the group $D\mu/T$ is independent of temperature.

The dispersion coefficient is influenced primarily by variations in the molecular diffusion coefficient. For the isothermal film, the dispersion coefficient is relatively independent of the shear stress at the gas-liquid interface. For systems in which $D\mu/T$ is independent of temperature, a larger temperature drop through the film drives the film toward plug flow. Also, the higher the interfacial shear, the more the film is driven away from plug flow.

For liquid films with low Reynolds numbers and predetermined film thickness, this analysis is expected to allow a test to be made in order to determine whether or not the film is in laminar flow. Conversely, if the film is known to be in laminar flow, this analysis permits the film thickness to be determined.

NOTATION

A = $(\Delta^2 U_1/D_0)\ \partial C_0/\partial X_*$
C = concentration of tracer normalized to initial concentration
D = molecular diffusion coefficient of tracer
F = F diagram as defined by Equation (11)
g = acceleration due to gravity
I = function as defined by Equation (30)
k = virtual coefficient of diffusion as defined by Equation (28)
L = downstream position where film is sampled for tracer content
M = constant as defined by Equation (13)
N = constant as defined by Equation (13)
ΔP = pressure drop
Q = volume of tracer of unit concentration which would correspond to the actual amount of tracer introduced into the fluid
Q_* = tracer flux as defined by Equation (24)
R = distance from tube center to gas-liquid interface
T = temperature
t = time
t_0 = breakthrough time as defined by Equation (36)
U = velocity
$<U>$ = average film velocity
V = total volume of liquid in reactor
x = distance downstream of tracer injection
X_* = $x - <U>t$
y = distance from wall to point within film

Greek Letters

α = $-N(T_1 - T_0)/T_0^2$
β = $\Delta/\mu\ (\tau_1/U_1)(e^\alpha - 1)/\alpha$
Δ = film thickness
$\delta(x)$ = delta function
ρ = liquid density
η = y/Δ
η_* = solution to Equation (35)
τ = shear stress
θ = angle as defined in Figure 1
μ = viscosity of liquid
Γ = volumetric flow rate per wetted perimeter

Subscripts

1 = gas-liquid interface
0 = wall (except where specified)

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APPENDIX A

Consider the laminar Couette flow of a liquid film in which $\mu = \mu_0 e^{-\alpha\eta}$. Furthermore, let us consider the case in which the binary diffusion coefficient is constant and equal to $D = D_0$. To avoid confusion, this case will be called the hypothetical case. Then, the following relationship may be easily derived:

$$\frac{U}{U_1} = \frac{e^{\alpha\eta} - 1}{e^\alpha - 1} \tag{1A}$$

$$\frac{<U>}{U_1} = \frac{e^\alpha - \alpha - 1}{\alpha e^\alpha - \alpha} \tag{2A}$$

and

$$C = C(0) + A\left[\frac{e^{\alpha\eta}}{\alpha} - \frac{\eta^2 e^\alpha}{2} + \frac{\eta^2}{2} - \eta - \frac{1}{\alpha}\right] \tag{3A}$$

Following the analysis described previously

$$I(\alpha, 1) = \frac{(2\alpha^2 - 9\alpha + 12)e^{2\alpha} + (2\alpha^2 - 24)e^\alpha + (2\alpha^2 + 9\alpha + 12)}{6\alpha^4 (e^\alpha - 1)^2} \tag{4A}$$

As expected, when $\alpha \to 0$, $I \to 1/120$. The dispersion coefficient is again given by Equation (29). For large values of α the dispersion coefficient for the hypothetical case may differ greatly from the Stokes-Einstein liquid (see Figure 5). The residence time distribution function is given by Equation (34).

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