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Gas Absorption by a Liquid Layer Flowing on the Wall of a Pipe

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A method is presented for predicting absorption rates for liquid layers flowing along a wall. It is based on results presently available in the literature as well as on the results of studies we recently carried out on the rate of oxygen absorption from a flowing air stream into water layers on the bottom of a horizontal enclosed channel and on the inside of a vertical pipe. Absorption measurements can be interpreted by assuming that the process is controlled by eddies whose length and velocity are characterized by bulk turbulence properties and that in a region of thickness δ close to the interface the turbulence is dampened by viscosity.

SCOPE

The absorption of a gas by a flowing liquid film, for which the primary resistance to mass transfer is in the liquid phases, is an important commercial process. Kamei and Oishi (1955), Lamourelle and Sandall (1972), Emmert and Pigford (1954), Menez and Sandall (1974), Stirba and Hurt (1955), Coeuret et al. (1970), Miller (1949), and Vyazovov (1940) have measured rates of ab-

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sorption for free falling water films. In spite of this large effort, no uniformly accepted method for the prediction of mass transfer rates has appeared.

The rate of absorption by a liquid film can be greatly increased by causing the gas to flow concurrently. Only limited information is available on the influence of gas flow. This includes the work by Chung and Mills (1974), Kasturi and Stepanek (1974), and Banerjee, Scott, and Rhodes (1970).

In this paper we present a method for predicting absorption rates for liquid layers flowing along a wall. It is based on results presently available in the literature as well as on the results of studies we recently carried out on the rates of absorption from a flowing air stream into water layers on the bottom of a horizontal enclosed channel and down the inside of a vertical pipe. The liquid Reynolds numbers in these experiments varied from 350 to 7 300 in the channel and from 280 to 10 600 in downflow. The range of gas Reynolds numbers was from 5 900 to 38 400 in the channel and from 3 500 to 34 100 in downflow. . .

CONCLUSIONS AND SIGNIFICANCE

For free falling films, we recommend use of the relation

$$Sh_m Sc^{-1/2} = 0.0077\, m^{+3/2}$$

to predict the liquid mass transfer coefficient k_L. In this equation

$$m^+ = \frac{mv^*}{\nu_L}$$

is the dimensionless thickness of the film, and v^* is a characteristic friction velocity

$$v^* = \sqrt{\frac{\tau_c}{\rho_L}}$$

where $\tau_c = 2/3\, \tau_w + 1/3\, \tau_i$.

For free falling films, $\tau_c = 2/3\, \tau_w$, and m^+ is related to the film Reynolds number by

$$m^+ = [(0.707\, Re_{LF}^{0.50})^5 + (0.0310\, Re_{LF}^{0.90})^5]^{0.20} = \gamma'$$

For sheared liquid films, m^+ is related to film Reynolds number by

$$m^+ = [(0.707\, Re_{LF}^{0.50})^{2.5} + (0.0379\, Re_{LF}^{0.90})^{2.5}]^{0.40} = \gamma$$

It is found that under some circumstances this relation can underestimate the value of Sh_m for sheared films by as much as a factor of three and so is a conservative estimate. The effect of gas flow can be better taken into account by the relation

$$Sh_m Sc^{-1/2} = 0.0077\, \gamma^{3/2}[1 + 1.8 \exp(-30F)]$$

where

$$F \equiv \frac{\gamma}{Re_G^{0.90}} \frac{\nu_L}{\nu_G} \sqrt{\frac{\rho_L}{\rho_G}}$$

Presently available models of the convective motions close to an interface are not consistent with the observed influence of liquid and gas flow on the absorptoin rate. We interpret absorption measurements by assuming that the process is controlled by eddies whose length and velocity are characterized by bulk turbulence and that in a region of thickness δ close to the interface the turbulence is dampened by viscosity. Presently available data suggest that this region is characterized by the relation $\delta v^*/\nu =$ constant.

For liquid phase controlled absorption, the liquid at the interface is saturated with the solute. The rate of absorption per unit area N can then be characterized by a mass transfer coefficient k_L, defined by the equation

$$N = k_L(c^* - c_b) \tag{1}$$

An examination of the results for free falling films shows that the data can be correlated by an equation of the form

$$Sh_m Sc^{-1/2} = f(Re_{LF}) \tag{2}$$

Here, Sh_m is a Sherwood number using the thickness of the liquid layer as the length parameter $Sh_m = k_L m/D$, and Re_{LF} is the Reynolds number of the liquid layer $Re_{LF} = 4\Gamma/\nu_L$.

Our initial effort to correlate measured values of k_L for finite gas flow was to use the above equation derived for zero gas flow. According to this equation, the influence of an increasing gas velocity at a fixed liquid flow rate would appear as a decrease in the thickness of the liquid layer and a proportionate increase in k_L. It was found that under certain conditions the prediction is satisfactory; however, under other conditions, the prediction of k_L can be too low by up to a factor of three.

In order to take into account the influence of a gas flow, an additional dimensionless group F was introduced. This is essentially a modified Martinelli parameter

$$F = \frac{\gamma}{Re_G^{0.90}} \frac{\nu_L}{\nu_G} \sqrt{\frac{\rho_L}{\rho_G}} \tag{3}$$

$$\gamma = [(0.707\, Re_{LF}^{1/2})^{2.5} + (0.0379\, Re_{LF}^{0.90})^{2.5}]^{0.40} \tag{4}$$

This parameter has been used in a previous paper (Henstock and Hanratty, 1976) to correlate measurements of film height and interfacial drag for annular two phase flows.

The magnitudes of measured mass transfer coefficients are much larger in virtually all cases than would be expected for laminar flow conditions. The convective motions in the bulk fluid controlling mass transfer, characterized by a velocity v_o and a length l_o, experience spatial variation over a distance δ from the interface. We find that assumptions in the literature, either explicit or implicit, regarding l_o, v_o, and δ are not consistent with the measurements discussed in this paper. Consequently, we explore a model whereby v_o and l_o characterize large scale disturbances in the bulk, and δ characterizes a thin region close to the interface where the convective motions are highly damped by viscosity.

THEORY

Momentum Transport in Wall Layers

Considerable success has been experienced in deriving an expression for the thickness of a wall layer by assuming that the variation of the eddy viscosity ν_T is the same as what would be experienced by a single phase flow. The shear stress is related to the velocity gradient through the relation

$$\frac{\tau}{\rho} = (\nu + \nu_T)\frac{d\bar{U}}{dy} \tag{5}$$

Henstock and Hanratty (1976) used the van Driest equation to estimate ν_T and obtained the following relations for sheared and for freely falling wall layers on the inside wall of a circular pipe under fully developed conditions, respectively:

$$m^+ = [(0.707\, Re_{LF}{}^{0.5})^{2.5} + (0.0379\, Re_{LF}{}^{0.90})^{2.5}]^{0.40} \tag{6}$$

$$m^+ = [(0.707\, Re_{LF}{}^{0.5})^{5} + (0.031\, Re_{LF}{}^{0.90})^{5}]^{0.20} \tag{7}$$

The friction velocity in the above relation is defined as

$$v^* = \sqrt{\frac{\tau_c}{\rho}} \tag{8}$$

where τ_c is an average shear stress in the layer defined as

$$\tau_c = \tau_w\left(1 - \frac{2}{3}\frac{m}{d_t}\right) - \frac{1}{3}\rho_L m\left[\frac{1}{\rho_L}\left|\frac{dp}{dz}\right| + g\right] \tag{9}$$

For $m/d_t \to 0$

$$\tau_c = \frac{2}{3}\tau_w + \frac{1}{3}\tau_i \tag{10}$$

For free falling liquid layers

$$\tau_c = \sqrt{\frac{2}{3}\rho_L g m} \tag{11}$$

and

$$m^+ = \frac{m^{3/2}\sqrt{\frac{2}{3}g}}{\nu} \tag{12}$$

Equation (6) is found to fit experimental data for sheared liquid layers covering a Reynolds number range of $20 < Re_{LF} < 16\,000$. Equation (7) for free falling layers agrees with the relations developed by Brauer (1956) and by Feind (1960) at large Re_{LF} and with Nusselt's (1916) relation at small Re_{LF}.

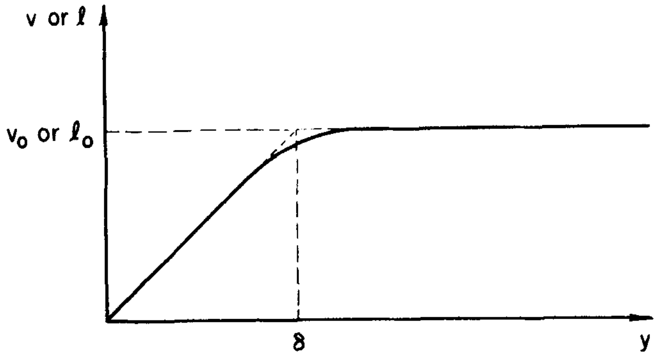

Fig. 1. Fluctuating velocity and mixing length profiles.

Prediction of the Height of the Wall Layer

Equation (6) is inconvenient to calculate the height of a sheared wall layer since the shear stress at the interface τ_i needs to be known.

Henstock and Hanratty (1976) have developed a relation between the friction factor characterizing the gas flow over the roughened liquid interface and the dimensionless group F defined in the introduction. By using this relation to eliminate τ_i from (6), the following more useful equations were developed for the height of a sheared wall layer under fully developed conditions:

$$\frac{m}{d_t} = \frac{6.59\,F}{(1 + 1400\,F)^{1/2}} \tag{13}$$

for vertical flows.

Motivation for the Method of Analysis

The interpretation of the mass transfer data has been motivated by the success which Henstock and Hanratty (1976) have experienced in using eddy viscosity concepts to describe momentum transfer in wall films over the range of liquid Reynolds numbers considered in this paper. The basic concept is to scale the convective motions responsible for momentum transfer in the same way as has been done for single phase turbulent flow in a channel or pipe. Such an approach has been used quite successfully to describe heat transfer in liquid films (Davis, Hung, and Arciero, 1975; Rosson and Meyers, 1965; Dukler, 1959; Hewitt, 1961). Consequently, in evaluating different theoretical approaches for describing gas absorption, we assume that the length and velocity of the convective motions close to the interface scale the same as for single phase turbulent flows.

Furthermore, the successful treatment of momentum transfer and heat transfer without taking special account of the large wave structure suggests that this also be done in correlating mass transfer data. A number of experiments support this approach. For concurrent flow in a horizontal channel, there is a large range of flow conditions for which no roll waves exist. For the experimental data reported in this paper, the same correlation for film height and for mass transfer coefficients held regardless of whether roll waves exist and no dramatic change in height or in mass transfer rate was noted at the transition to roll waves. Similar results have been obtained by Davis (1975) in his heat transfer studies. This approach implies that the convective motions at different locations in the film are approximated by making a pseudo steady state assumption. It also implies that if the convective motions responsible for heat, mass, and momentum transfer are controlled by wave properties, then these should be characterized by the small waves riding on the roll waves and not by the parameters describing the roll waves.

Using the viewpoints expressed above, we could not find any theory in the literature capable of interpreting our mass transfer results. We have been lead to the conclusion that a basic failing of all of these theories is that they use a single length scale to characterize the convective motions. We suggest that at least two length scales are needed, one to characterize the convective motions l_o and another to characterize the thickness of a damped region close to the interface δ.

A Model for the Mass Transfer Process

In an analogous way to momentum transport, convective mass transport can be characterized by an eddy diffusion coefficient D_t. The rate of mass transferred per unit area is given as

$$N = -(D + D_t)\frac{dc}{dy} \tag{14}$$

where D_t is a function of y. The form of D_t is usually taken to be

$$D_t = ay^n \tag{15}$$

Son and Hanratty (1967) have shown that for this case the resulting mass transfer coefficient is

$$k_L = \frac{n}{\pi} a^{1/n} \sin\left(\frac{\pi}{n}\right) D^{\left(\frac{n-1}{n}\right)} \tag{16}$$

This particular form of D_t is sometimes justified as being the first term in a Taylor series expansion of a more complicated function of y. In the region of interest near the interface, y is small, and only the first term is necessary.

The mass transfer process may be pictured as occurring in much the same way as the momentum transfer process described by Prandtl in his formulation of the mixing length model. A discrete packet of fluid moves a distance l in the y direction, with a velocity v, to a position where the bulk concentration differs by an amount Δc. The effective diffusivity (or eddy diffusivity) resulting from such a transfer can be shown to be

$$D_t \sim vl \tag{17}$$

The velocity v is taken to be given by the root mean square of the velocity fluctuations in the y direction; l is a mixing length characterizing the mass transfer process.

These two scales will be functions of y. Based on measurements in single phase flows, their values in the bulk of the flow can be expected to be approximately constant $v = v_o$ and $l = l_o$. Across some region near the interface with a thickness δ, they will be damped from their bulk value to zero. Considerations of continuity suggest that the damping of v very near the interface should be linear. By analogy to momentum transfer, the damping of l may also be taken to be linear. The profiles of v and l would then be as given in Figure 1. For high Schmidt numbers, the concentration boundary layer will be much thinner than δ, and the limiting value of v and l close to the interface can be used

$$v \sim \frac{v_o}{\delta} y \tag{18}$$

and

$$l \sim \frac{l_o}{\delta} y \tag{19}$$

The resulting form for D_t is then

$$D_t \sim \frac{v_o l_o}{\delta^2} y^2 \tag{20}$$

Thus, parameter a is directly proportional to $v_o l_o/\delta^2$, and

$$k_L \sim \left(\frac{v_o l_o D}{\delta^2}\right)^{1/2} \tag{21}$$

Note that this formulation is given in terms of three independent parameters: v_o, l_o, and δ. An alternative formulation would be to use the length scale δ, velocity scale v_o, and a time scale

$$\tau = l_o/v_o \tag{22}$$

The expression for k_L then becomes

$$k_L \sim \left(\frac{v_o^2 \tau D}{\delta^2}\right)^{1/2} \tag{23}$$

This is not a fundamentally different formulation, but it is merely defined in terms of different variables.

We now exploit the assumption that convective motions in the fluid can be characterized in the same way as turbulence properties for a single phase flow. The scales v_o and l_o then characterize the turbulent viscosity predicted by a single phase relation, such as the van Driest equation, at a distance from the wall equal to m. This type of argument supports the choices of

$$l_o \sim m \tag{24}$$

$$v_o \sim v^* \tag{25}$$

for the characteristic length and velocity of the convective motions close to the interface. A comparison of the correlations developed from suggested models (Table 1) with the measurements for freely falling layers indicates that none of them properly takes into account the influence of liquid Reynolds number.

Consequently, we explored a method for estimating the thickness of the region over which the convective motion is dampened, δ, that had not previously been suggested. If this damping is due completely to viscosity, then

$$\delta = f(v_o, l_o, \nu) \tag{26}$$

The results on free falling layers would be consistent with the above interpretation if δ is assumed independent of l_o. Then

$$\frac{\delta v_o}{\nu} = \text{constant} \tag{27}$$

If (25), (26), and (28) are substituted into (22), the relation for the mass transfer coefficient becomes

$$k_L \sim \left(\frac{v^{*3} m D}{\nu^2}\right)^{1/2} \tag{28}$$

In dimensionless terms

TABLE 1. COMPARISON OF MASS TRANSFER THEORIES

Worker	l_o	v_o	τ	δ	D_t	$Sh_m\, Sc^{-1/2}$
Levich, Davies	—	v^*	δ/v_o	$\frac{\sigma}{\rho v_o^2}$	$\frac{\rho v^{*3}}{\sigma} y^2$	$Ca^{1/2}\, m^+$
Banerjee, Scott, and Rhodes	a	$\left(\frac{a}{\lambda}\right) c$	—	l_o	$\frac{a^2 c}{\lambda^3} y^2$	$Re_{LF}^{1/2}$
Lamont and Scott; King	l_k	v_k	—	l_o	$\frac{\epsilon^{1/2}}{\nu} y^2$	$m^{+\,3/4}$
Fortescue and Pearson	Macroscale	Intensity	—	l_o	$\frac{v_o}{l_o} y^2$	$\frac{m^2 v_o^{1/2}}{l_o \nu}$
This work	m	v^*	—	$\frac{\nu}{v^*}$	$\frac{m v^{*3}}{\nu^2}$	$m^{+3/2}$

$$Sh_m \sim \left(\frac{mv^*}{\nu} \right)^{3/2} Sc^{1/2} \qquad (29)$$

The group mv^*/ν is a dimensionless film thickness usually denoted as m^+, so that

$$Sh_m \sim m^{+3/2} Sc^{1/2} \qquad (30)$$

Other Models for the Mass Transfer Process

A comparison of this method of interpretation with other approaches is summarized in Table 1. The surface renewal models evidently implicitly assume that $\delta \sim l_o$. In terms of the characteristic time τ defined above

$$k_L \sim \left(\frac{D}{\tau} \right)^{1/2} \qquad (31)$$

This time τ may therefore be associated with the average surface age, or the rate of surface renewal. No methods for applying the surface renewal models are usually given.

Fortescue and Pearson (1967), Lamont and Scott (1970), and Banerjee, Scott, and Rhodes (1970) model the convective motions as arrays of roll cells at the liquid surface. All are equivalent to using an eddy diffusivity model, with δ taken to be equal to l_o, and with v_o and l_o as the characteristic velocities and lengths of the eddies. Fortescue and Pearson estimated v_o and l_o from the macroscales of the flow, while the others took them to be given by the scales of the dissipative eddies. King (1966) developed his model in terms of an eddy diffusivity, but it appears to be equivalent to the small roll cell model of Lamont and Scott.

A number of authors have considered the influence of regular waves on mass transfer (Ruckenstein and Berbente, 1968; Javdani, 1974). These analyses predict a much smaller enhancement of the mass transfer rate than is observed in the experiments discussed in the paper and are applicable to films flowing at lower Reynolds numbers. Banerjee, Rhodes, and Scott (1967) considered the Reynolds number range of interest in this paper by describing convective motion near the interface as cellular motions due to irregular waves. According to this model, $l_o \sim (a/\lambda)\lambda$, $v_o \sim a/\lambda\ c$, and $\delta \sim 1$.

Levich (1962) and Davies (1972) assume that the damping near the interface is due to surface tension, and so

$$\delta \sim \frac{\sigma}{\rho v_o^2} \qquad (32)$$

In turn, they take $l_o = \delta$, but v_o is assumed to be given by the friction velocity v^*.

In order to compare these models, it is necessary to estimate the Kolmogorof scales of the dissipative eddies and the wave parameters. The former are given by

$$l_k = \left(\frac{\nu^3}{\epsilon} \right)^{1/4} \qquad (33)$$

and

$$v_k = (\nu\epsilon)^{1/4} \qquad (34)$$

The dissipation term can be estimated by

$$\epsilon \sim \frac{v_o^3}{l_o} \qquad (35)$$

as suggested by Brumfield, Houze, and Theofanous (1975). The wavelength is estimated by $\lambda \sim m$ and $c \sim U_L$. The forms for k_L and Sh_m which result from these estimates are shown in Table 1. While somewhat different ways of estimating ϵ, λ, and c are possible, the results would not be too different.

This table points out that the models used are basically similar. The apparent differences arise from three sources. First, some authors work with a characteristic time rather than a length. Second, most take $\delta = l_o$, limiting the number of independent parameters. Third, the methods of application differ, depending on whether large eddy motions, small eddy motions, or wave motions are thought to be controlling the mass transfer process. All the theories predict dependence on diffusivity to the one half power, so the result cannot be differentiated on that point. However, the predicted dependence of Sh_m on m^+ is quite different, with the largest predicted dependence being the method proposed in this paper.

Brumfield, Houze, and Theofanous (1975) have recognized in a direct way the intermittent nature of the flow in falling films in their interpretation of mass transfer results. They used a Fortescue and Pearson model to describe mass transfer to the roll waves, assuming $v_o = v^*$ and $l_o/m = 2\,000/Re$. Mass transfer to the base film was described by a surface renewal theory, where the renewal rate is related to the velocity and spacing of the roll waves. They used wave parameters measured by Telles and Dukler (1970) to calculate mass transfer rates. Their predictions agree with measurements for $1\,200 < Re < 3\,800$ but are higher at $Re = 5\,700$. We did not use a similar approach because of the reasons cited previously. In addition, the physical justification for the assumed relation for l_o and for ignoring the influence of convective motions in the base film is not apparent to us.

EXPERIMENTAL

Mass transfer coefficients were measured for two different flow configurations. One of these was a horizontal rectangular channel, which has been described in detail by Miya (1970). The channel was 1 in. high by 12 in. wide and approximately 32 ft long. The liquid flowed along the bottom wall, and air flowed concurrently above the liquid. The drag of the air on the liquid provided the force for motion of the film. For mass transfer studies, the liquid used was taken from the house water supplies, run once through, and discarded. Flow rates were measured with one of two calibrated rotameters. Air was supplied by a large blower and flow rates measured by a series of calibrated orifice plates.

The other flow loop used had a vertical inch diameter pipe approximately 30 ft long. The air and water flowed concurrently downward. Again, house water was run once through and discarded, and flow rates were measured with two calibrated rotameters. Air was supplied by another blower and flow rates measured with a series of calibrated orifice plates. The section of the equipment in which mass transfer measurements were made was fabricated from nominal 1 in. ID Plexiglas tubing. The liquid entered the pipe through an adjustable annular slot at a 30 deg angle to the direction of flow in order to minimize entry disturbances.

Mass transfer coefficients were measured in the same way in both experiments. Since the house water was found to be oxygen poor, the rate of oxygen absorption from air was measured. The liquid film was sampled at two points by inserting 18 gauge hypodermic needles through the equipment wall at a 45 deg angle to the direction of flow and facing into it. Samples were withdrawn continuously and run through epoxy cells containing Beckman polarographic oxygen electrodes. By throttling the sampling lines, it was possible to obtain air free samples with a minimum disturbance to the liquid film. To ensure that the samples were representative of the bulk concentration of the film, dye was injected through a very small hole in the equipment wall. Only those flow conditions for which the dye mixed completely with the film within a few inches were studied. In addition, the sampling needles were traversed as far as possible through the thickness of the film without breaking the surface for a few cases. No measurable concentration gradient was found, indicating the film was well mixed.

By measuring the change in concentration between two points, the flow rates and the distance between the points, a

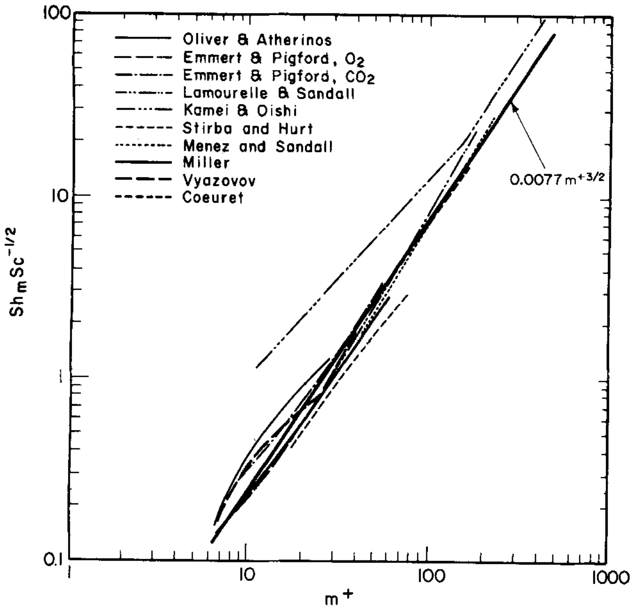

Fig. 2. Sh_m as a function of m^+, free-falling film data.

mass transfer coefficient could be calculated. The positions of the sampling probes were picked such that a significant change in concentration occurred, but the downstream concentration did not too closely approach saturation. Within these constraints, the positions of the probes were varied as widely as possible to check for effects of length. The area for mass transfer was taken as that for a smooth film. Thus, the reported mass transfer coefficients include some effect of area increase.

CORRELATION OF RESULTS FOR FREE FALLING FILMS

The data for free falling liquid films are shown in Figure 2, plotted as $Sh_m Sc^{-1/2}$ vs. m^+. The range of variables covered in these investigations is summarized in Table 2. The value of the film thickness m was calculated as outlined above. The friction velocity used is the characteristic friction velocity developed from consideration of two phase flows. For free falling films, it is given as

$$v_c^* = \sqrt{\frac{2}{3}\frac{\tau_w}{\rho_L}} \tag{36}$$

so that m^+ is given by $m^+ = \gamma' (Re_{LF})$, again as outlined above. For clarity, each set of data is represented by a single line so the actual scatter in the data is considerably more than it might appear.

Considerable differences among the workers are noted. However, over the range of m^+ covered, it appears that the data can be represented well by

$$Sh_m Sc^{-1/2} = 0.0077\, m^{+3/2} \tag{37}$$

This is indicated by the solid, heavy line in the figure. The data of Kamei and Oishi appear to be higher than the rest at lower values of the Reynolds number. A possible explanation for this will be discussed below.

Note that because m^+ is a unique function of Re_{LF}, this relation can be written in terms of the more commonly used variables as

$$Sh_m Sc^{-1/2} = 0.0077\, \gamma'^{3/2} \tag{38}$$

If all terms in Re_{LF} are collected to one side, this takes the form

$$\frac{k_L\, \nu_L^{1/6}}{g^{1/3}\, D^{1/3}} = 0.0067\, \gamma'^{5/6} \tag{39}$$

This rather odd function of Reynolds number is a curved line, even when plotted on logarithmic paper.

The observation that the bulk of the data are well represented by a relation proportional to $m^{+3/2}$ lends support to the ideas that large scale motions are controlling in the mass transfer process and that δ is given by Equation (28).

INFLUENCE OF GAS VELOCITY

Our measurements of mass transfer coefficients for free falling films are shown in Figure 3. The curve represents Equation (37), developed from data in the literature. The measurements agree quite well with this correlation. This agreement indicates that our measurement technique is accurate.

The measurements for concurrent downward flow and for concurrent flow in the channel are shown in Figures 4 and 5, respectively. The predicted values of Sh_m are somewhat different for these cases than for free falling films because the relation between m^+ and Re_{LF} for concurrent flow is

$$m^+ = [(0.707\, Re_{LF}^{1/2})^{2.5} + (0.0379\, Re_{LF}^{0.90})^{2.5}]^{0.40} \tag{40}$$

Thus, the predicted relation for Sh_m for this case is

$$Sh_m Sc^{-1/2} = 0.0077 \gamma^{1.5} \tag{41}$$

which predicts somewhat larger values of Sh_m than the free falling case. Equation (41) is shown in Figures 4 and 5.

It can be seen that all of the measurements of Sh_m with a concurrent gas flow are as large as or larger than predicted by Equation (41). This same type of behavior has also been noted in the data of Chung and Mills (1974) and of Kasturi and Stepanek (1974). See, for example, Figure 6. Equation (41), therefore, represents a conservative estimate of the values of k_L for systems with gas flow.

Table 2. Experimental Conditions, Mass Transfer Results

Worker	Flow configuration	Length	Solute	Range of Re_{LF}
Kamei and Oishi	4.76 cm pipe	250 cm	CO_2	300-30 000
Oliver and Atherinos	Channel at 7.5 deg slope	96.5 cm	CO_2	100-1 200
Lamourelle and Sandall	Outside of 1.59 cm rod	91.5-183 cm	O_2, H_2, He	1 300-8 000
Emmert and Pigford	2.54 cm pipe	114 cm	O_2, CO_2	250-1 000
Stirba and Hurt	2.16 cm and 2.54 cm pipes	91.5 cm, 194 cm	CO_2	280-3 750
Menez and Sandall	Outside of 1.59 cm rod	183 cm	CO_2	2 100-10 000
Coeuret et al.	0.4 cm pipe	Not reported	CO_2	700-9 000
Miller	Not reported	73.2-116 cm	O_2	100-3 000
Vyazovov	Channel at 17.5 deg	37.5-73.2 cm	CO_2	100-900
This work	2.54 cm pipe	90-210 cm	O_2	340-10 900

All workers used water as the liquid phase.

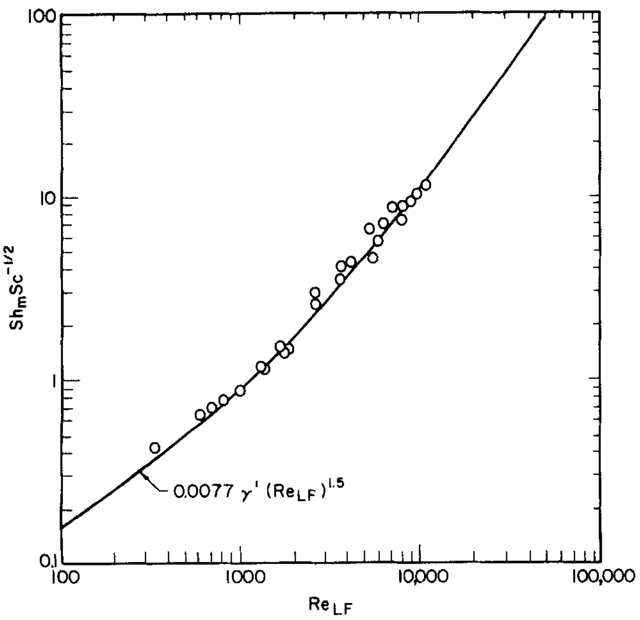

Fig. 3. Oxygen transfer data for free-falling films.

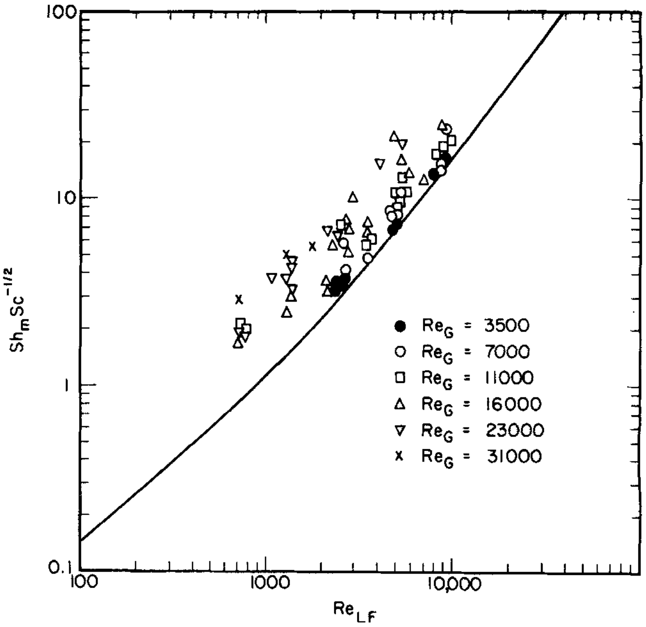

Fig. 4. Concurrent mass transfer data, downflow.

The measurements shown in Figures 4 and 5 indicate that the mass transfer coefficients approach the value predicted from consideration of free falling films with decreasing gas Reynolds number or increasing liquid Reynolds number. Since the dimensionless group F varies roughly as the ratio of liquid to gas flow rates, this suggests itself as an appropriate variable with which to correlate the increase in Sh_m caused by the presence of a gas flow. This is shown in Figures 6 and 7 for vertical flows and for flow in the horizontal channel, respectively. The data for Chung and Mills and of Kasturi and Stepanek are also shown in Figure 5. Here the ordinate I is the ratio of the measured values of Sh_m for finite gas flows to that which would be predicted from Equation (41). At large F, the ratio I approaches a value of unity, while at small F it appears to approach a value of approximately 2.8. These results can be correlated by the following empirical fit to the data:

$$I = 1 + 1.8 \exp(-30F) \qquad (42)$$

or

$$Sh_mSc^{-1/2} = 0.0077^{1.5}[1 + 1.8 \exp(-30F)] \qquad (43)$$

It should be emphasized that this correlation is empirical and that the choice of functional dependency on F has no theoretical foundation.

For a given flow configuration, and for high velocity gas flows, the ratio of film thickness to pipe diameter is a function of only F. Equation (43) can then be put in a somewhat more convenient form by dividing by m/d. Then

$$Sh_dSc^{-1/2} = 0.0077\gamma^{1.5}\left[\frac{1 + 1.8\exp(-30F)}{m/d}\right] \qquad (44)$$

where $Sh_d = k_L d_t/D$. For vertical flow in a pipe, for instance

$$\frac{m}{d_t} = \frac{6.59\,F}{\sqrt{1 + 1\,400\,F}} \qquad (45)$$

and

$$Sh_dSc^{-1/2} = 0.0077^{1.5}\left[\frac{[1 + 1.8\exp(-30F)]\sqrt{1 + 1\,400\,F}}{6.59}\right] \qquad (46)$$

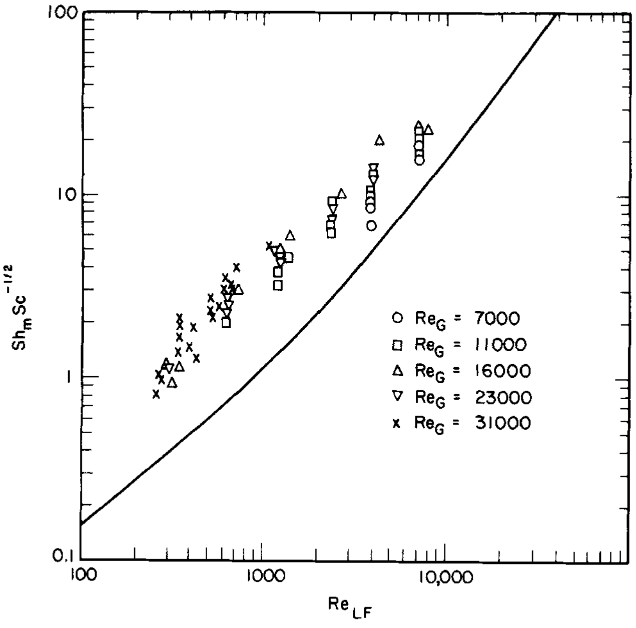

Fig. 5. Concurrent mass transfer data, horizontal channel.

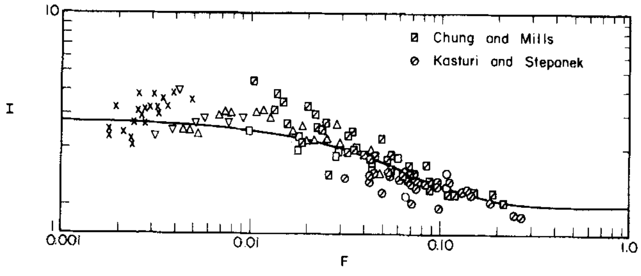

Fig. 6. Increase in Sh_m as a function of F, vertical flows, including data of Chung and Mills, Kasturi and Stepanek.

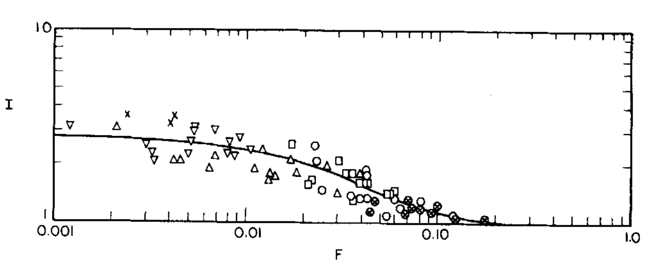

Fig. 7. Increase in Sh_m as a function of F, horizontal channel.

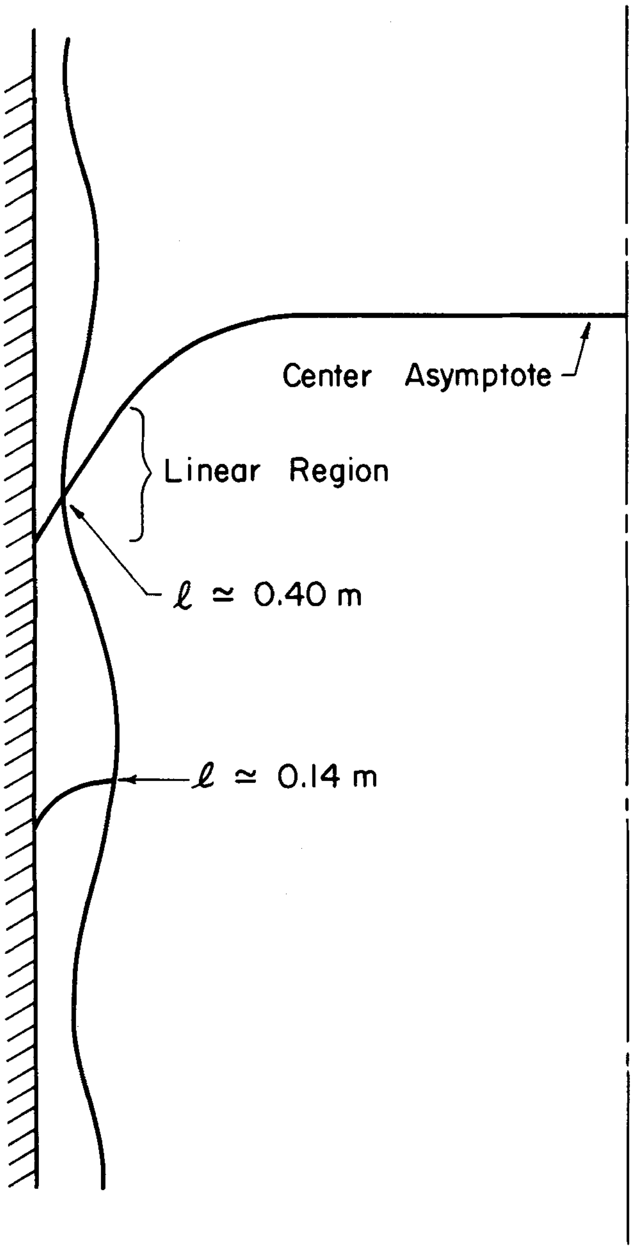

Fig. 8. Mixing length profiles.

$$= 0.0077^{1.5}\, g(F) \qquad (47)$$

INTERPRETATION OF RESULTS ON THE INFLUENCE OF GAS VELOCITY

According to the theory developed above, the influence of a concurrent gas flow would be felt through its influence on v_o and l_o. Thus, for a given liquid Reynolds number, an increase in gas flow rate should cause a decrease in the film thickness m and a proportionate increase in the mass transfer coefficient k_L, keeping the value of Sh_m constant. This was found experimentally to be only approximately the case, with k_L increasing somewhat more rapidly than m decreased.

A possible reason for this discrepancy may be the way that l_o and v_o are scaled with bulk flow parameters. In this analysis, it was assumed that l_o was related to m, and v_o was related to v^* by constant proportionality factors. Now, from measurements in single phase flows, it is known that the mixing length near the wall is proportional to distance from the wall, the proportionality given by von Karman's constant. In the center of the pipe the mixing length is approximately constant; Schlichting (1968) indicates (p. 569) that it reaches a value of about 0.14 times the pipe radius. If a similar profile exists entirely within the film, the value of l_o would be approximately 0.14 times the film thickness. This might be expected to hold for free falling films, which would not see any effect of the gas core.

For a high velocity gas flow, however, the mixing length profile must extend across the entire pipe. Since the linear region extends out to about one tenth of the diameter, all liquid films would be completely in that portion of the profile. The value of l_o would then be roughly 0.40 times the film thickness. In both cases, l_o is proportional to m, but proportional by different factors. This is illustrated in Figure 8.

Similarly, measurements of the y component of the fluctuating velocity presented by Hinze (1959, p. 521) show that near the center of the pipe $v_o \simeq 0.03\, v^*$, whereas near the wall $v_o \simeq 0.04\, v^*$. Thus, v_o would also scale with v^* somewhat differently for free falling and highly sheared films. Since

$$k_L \sim \left(\frac{v_o^3 l_o}{\nu^2}\right)^{1/2} \qquad (48)$$

for δ given by ν/v_o, the expected change in k_L would be of the order of

$$\left(\frac{0.04}{0.03}\right)^{3/2} \left(\frac{0.40}{0.14}\right)^{1/2} = 2.6$$

This is approximately what is found. In addition, this analysis predicts that the effect of gas flow rate should reach an upper limit and not increase indefinitely, which is also found.

INFLUENCE OF FLUID PROPERTIES AND OF ATOMIZATION

It should be emphasized that all of the data analyzed in this paper were obtained using water as the liquid phase and were under conditions of little or no atomization. Although, as reported in the thesis by Henstock (1977), small rates of atomization have no significant effect on the mass transfer rate, there might be an effect at high rates due to associated disturbances of the liquid layer. Results with other liquids and for higher rates of atomization are needed in order to test the generality of the proposed correlation.

One of the reviewers of this paper has called to our attention the recent work by Chung and Mills (1974) in which the influence of liquid viscosity on the absorption of carbon dioxide was examined. The viscosity was varied twofold by varying the temperature of water and threefold by adding glycol to water. The comparison of these results with the correlation proposed in this paper is inconclusive. Good agreement is obtained with the water data at different temperatures except $Re_{LF} < 90$, where their results appear higher than those of other investigators. As with their water data, no significant effect of viscosity variation is noted in the runs with glycol solutions, if plotted as $Sh\, Sc^{-1/2}$ vs. Re_{LF}. However, in these runs the mass transfer measurements could be 50% higher than the proposed correlation at large Reynolds numbers. This was true even for the experiment with only 5% ethylene glycol for which the kinematic viscosity is 1.30×10^{-6} m^2/s, and the surface tension is very close to that for water.

CLOSURE

This paper presents a method for predicting the effect of gas flow on absorption into a wall layer. It gives a general correlation of data for water films both with and without concurrent air flow. In deriving this, we found that none of the methods of correlation proposed in the literature were adequate. This comparison leads us to the conclusion that all present theories are lacking because they use only a single length or time scale. At least two length scales will be needed. We use a method to correlate the data which uses separate length scales to characterize the disturbances close to the interface and the region of damping close to the interface. The theoretical implications of the method used to correlate our data could be more interesting than our presentation of a much improved design relation. Consequently, some further discussion of this theory seems appropriate.

The convection motions controlling absorption could be associated with irregular waves at the interface, or due to turbulence generated by the shear layers close to the wall. We have been encouraged to characterize them over a wide range of Reynolds numbers by using turbulence concepts for single phase flow because of the success of this approach in characterizing momentum transfer. However, we recognize that the interpretation which we presented for presently available results is probably not unique. In fact, presently available measurements cannot distinguish whether waves or turbulence are the controlling factors so long as the existence of a damped region close to the interface is recognized.

The suggestion of the existence of damping close to an interface is not novel. Literature dating back to 1930 can be cited which suggests the importance of such a region in controlling absorption rates (Davis and Crandall, 1930; Kanwisher, 1963; Boling, 1960; Kraus, 1972). It would appear noncontroversial that such a region exists. Its importance would depend on whether the thickness of the concentration boundary layer δ_c is of the order of or less than δ. From the data shown in Figure 3, we estimate for $Sc = 900$ that $\delta_c/m = 1/30$ at $Re_{LF} = 1\,000$ and that $\delta_c/m = 1/300$ at $Re_{LF} = 10\,000$. Consequently, it appears that (30) cannot be used at Reynolds numbers much smaller than investigated in this research, since the estimated δ_c/m would be much too large for the proposed theory to be valid.

At present, no theory exists to estimate δ, mainly because the kinematics of the convective motions close to the interface are not known. In general, one would expect that $\delta = f(v_o, l_o, \nu$, interfacial properties). If the kinematics were understood, the equations describing the velocity field close to the interface could be written. These equations would have viscous terms whose importance would vary with distance from the interface but would include no terms involving interfacial properties. These would appear in the solution of these equations through the boundary conditions. Since viscosity would be responsible for the actual damping, we expect this to be the primary effect. Consequently, in estimating δ we have initially ignored the influence of surface properties.

Another problem with the theory which is of some concern to us is the use of a mass transfer-momentum transfer analogy. Although a relation between eddy diffusivity and eddy viscosity is never formally stated, the eddy diffusivity is based on a characteristic velocity and a characteristic length which have been found to represent momentum transfer. The scales which best characterize mass transfer need not be the same as for momentum transfer, since they could be related to the structure of the concentration field as well as the velocity field.

ACKNOWLEDGMENT

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NOTATION

a_{LF} = area liquid film per unit length
A_t = tube area
c = concentration of solute; wave velocity
c^* = concentration of solute at the interface
c_b = bulk concentration of solute
D = mass diffusivity
D_t = turbulent diffusivity
d_t = tube diameter
F = dimensionless group characterizing two phase flow,

$$F \equiv \frac{\gamma}{Re_G{}^{0.90}} \frac{\nu_L}{\nu_G} \sqrt{\frac{\rho_L}{\rho_G}}$$

I = increase in Sh_m due to effect of cocurrent gas flow
k_L = mass transfer coefficient for the liquid phase
l = length scale of velocity fluctuations
l_o = length scale of velocity fluctuations in the bulk of the flow
m = film thickness
m^+ = dimensionless film thickness, $m^+ = mv^*/\nu_L$
N = mass flux of solute
Re_{LF} = Reynolds number of the liquid film, $Re_{LF} = 4\Gamma/\nu_L$
Sc = Schmidt number, $Sc = \nu_L/D$
Sh_m = Sherwood number based, $Sh_m = k_L m/D$
V_L = Superficial liquid velocity = $W_L/\rho_L A_t$
v = velocity fluctuation in the radial direction
v_o = velocity fluctuation in the bulk of the flow
v^* = friction velocity, $v^* = \sqrt{\tau_c/\rho_L}$
y = spatial coordinate normal to average direction of flow, $y = 0$, corresponds to the gas-liquid interface

Greek Letters

σ = surface tension
Γ = volumetric flow rate of liquid per unit of wetted perimeter
γ, γ' = functions of Re_{LF},

$$\gamma = [(0.707)Re_{LF}{}^{0.50})^{2.5} + (0.0379\,Re_{LF}{}^{0.90})^{2.5}]^{0.40}$$

$$\gamma' = [(0.707\,Re_{LF}{}^{0.50})^5 + (0.0310\,Re_{LF}{}^{0.90})^5]^{0.20}$$

δ = thickness of the region near the interface over which velocity fluctuations are damped
ϵ = rate of dissipation of turbulent energy per unit mass
ν_G = gas kinematic viscosity
λ = wavelength
ν_L = liquid kinematic viscosity
ν_t = coefficient of eddy kinematic viscosity
ρ_G = gas density
ρ_L = liquid density
τ = shear stress; also, time characteristic of velocity fluctuations
τ_c = characteristic shear stress:

$$\tau_c = 2/3\,\tau_w + 1/3\,\tau_i \quad \text{for} \quad m/d_t$$

τ_i = interfacial shear stress
τ_w = wall shear stress

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Residence Time Distributions of Paper Pulp Slurries in Vertical Laminar Flow

The hydrodynamic behavior of paper pulp slurries in vertical laminar flow has been observed and modeled. Radiotracers were used to tag both the liquid and solid phases of slurries flowing in 4 and 30 cm pipes. Residence time distributions were determined for each phase and were used to derive and compare several flow models. A model consisting of a central core in plug flow circumscribed by a clear Newtonian annulus provides a good representation of the observed slurry behavior. Correlations between the model parameters and the Reynolds number and slurry consistency are presented, and implications of the results on the design of slurry flow reactors are discussed.

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SCOPE

Modeling a slurry flow reactor involves the direct or indirect specification of the residence time distribution of the solid phase. Under different conditions, the slurry may be treated either as a homogeneous or a heterogeneous fluid; when the solid and liquid phases move together, the slurry can be dealt with as a single non-Newtonian liquid, while if slip occurs, the existence of separate phases must be explicitly taken into account.

The observation of a slurry flow using tracers is one way to determine how closely heterogeneous and homogeneous models approximate reality. Flow patterns can be observed by means of residence time distribution measurements; however, since the behavior of the phases may be different, each phase must be tagged separately.

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