Re = $D_p U_b \rho/\mu$ Reynolds number
U_b = bulk average velocity in pipe
U_o = velocity of fluid through orifice
β = D_o/D_p
σ = interfacial tension
μ = viscosity, with subscript for dispersed or continuous phase
τ_{crit} = stress marginally sufficient to fracture drop
ρ = density, with subscript for dispersed or continuous phase

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New Model for Turbulent Mass Transfer Near a Rigid Interface

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Mass transfer experiments at high Schmidt numbers can be used to study the space-time structure of the velocity field within the viscous sublayer. An interpretation of these experiments requires a theory, *albeit* approximate, which can relate the Stanton number St^+ to observable hydrodynamic parameters. In a series of papers (Petty, 1975; Petty and Wood, 1980a,b; Yao et al., 1981), we have been developing such a theory. So far, only qualitative agreement with experimental data has been possible, but this may only be due to a lack of accurate hydrodynamic parameters in the very near wall region (i.e., $x_1^+ < 5$). On the other hand, quantitative agreement between theory and experiments may require physical effects related to the nonlinear coupling between velocity and concentration fluctuations in the convective diffusion equation (see Shaw and Hanratty, 1977a,b; Yao et al., 1981).

In the present communication, a non-gradient model for the turbulent flux is derived from the elemental requirement that

$$\langle c'(\mathbf{x},t)\rangle = 0. \tag{1}$$

For a concentration field which is statistically stationary and statistically homogeneous in planes parallel to the mass transfer surface at $x_1 = 0$ (Figure 1), Eq. 1 implies that

$$\frac{d\langle u_1' c'\rangle}{dx_1} = -u_c(x_1)\frac{d\langle c\rangle}{dx_1} \tag{2}$$

for high Schmidt numbers. The "convective" velocity $u_c(x_1)$ depends on the underlying turbulent motion in the near wall region as well as the molecular diffusivity.

The specific objectives of this note will be to derive Eq. 2 and to show that a first-order approximation to $u_c(x_1)$ yields results for the mass transfer coefficient which are quantitatively consistent with experimental data for a range of hydrodynamic parameters estimated using recent data on bursting rates and turbulent intensities near rigid interfaces.

HYDRODYNAMICS IN THE NEAR-WALL REGION

Several authors have suggested that the near-wall region is inhabited by densely packed pairs of counter rotating vortices lying in the streamwise direction (cf., Blackwelder, 1979). There vortices are quite long in axial extent (greater than 1,000 ν/u^*) with a diameter of only about 30 ν/u^*. Periodically larger eddies from the outer region presumably induce an instability in the sublayer which causes a violent bursting or jetting of the sublayer fluid into the buffer zone. The mean period between these bursts is approximately $\langle \tau_M \rangle = 100\ \nu/u^{*2}$ (Berman, 1980). The probability distribution of τ_M is log normal and estimates of its variation are given in Nakagawa and Nezu (1978). Between the bursts the velocity auto-correlation is relaxing with a mean characteristic time, τ_H^+. For the high Schmidt numbers considered here, the resistance to mass transfer will be confined to a region near the wall that is much smaller in scale than the characteristic dimensions of the coherent sublayer structures. Thus the mass transfer rate will be limited by molecular diffusion and weak turbulent mixing caused by turbulent velocity fluctuations induced by the sublayer vortices.

Coles (1978) developed a sublayer model which can be used to estimate the intensity of $u_1'(x,t)$ in the near-wall region. The use of a model rather than actual data is necessary because for high Schmidt numbers the mean concentration field changes over distances within $x_1^+ \simeq 1$, which is well below the region probed in the thickest sublayer experiments of Kreplin and Eckelman (1979a). The model of Coles implies that $a_{11}^+ = 10^{-5}$, where $\langle u_1'^2\rangle = a_{11}x_1^4$.

A different approach for determining a_{11}^+ has recently been suggested by Campbell and Hanratty (1981). By assuming that the fluctuating velocity in the streamwise direction is nearly homogeneous (see the recent experimental measurements of Kreplin and Eckelmann, 1979b), Campbell and Hanratty used a two-dimensional continuity equation for the fluctuating velocity and experimental data on the spanwise velocity fluctuations to infer that $a_{11}^+ \simeq 10^{-5}$ in agreement with Coles' model.

MODEL FOR THE MEAN FIELD $\langle c\rangle(x_1)$

The convective-diffusion equation governs the instantaneous concentration field, i.e.,

$$\frac{\partial c}{\partial t} + \boldsymbol{u},\nabla c = \mathcal{D}\nabla^2 c. \tag{3}$$

For the problem defined by Figure 1, the mean field satisfies

$$\frac{d\langle u_1' c'\rangle}{dx_1} = \mathcal{D}\frac{d^2\langle c\rangle}{dx_1} \tag{4}$$

$$\langle c\rangle = c_w, \qquad x_1 = 0 \tag{5}$$

$$\langle c\rangle = 0, \qquad x_1 = \infty. \tag{6}$$

Equations 2, 4–6 can easily be solved for $\langle c\rangle(x_1)$. This result can then be used to calculate the average mass transfer coefficient or, equivalently, the Stanton number St^+.

The region within the viscous sublayer over which $\langle c\rangle(x_1)$ changes appreciably is $\delta_c \equiv D/\langle k_c\rangle$ (Figure 1). If the friction velocity u^* and the kinematic viscosity ν are used to introduce characteristic length and velocity scales, then $St^+Sc = 1/\delta_c^+$. The above boundary value problem gives the following expression for δ_c^+ in terms of Sc and $u_c^+(x_1)$

$$\delta_c^+ = \int_0^\infty d\eta \exp\left[-Sc\int_0^\eta u_c^+(\xi)d\xi\right] \tag{7}$$

CONSTITUTIVE MODEL FOR THE TURBULENT FLUX $\langle u_1' c'\rangle$

An evolution equation for the fluctuating concentration obtains by subtracting Eq. 4 from Eq. 3

$$\mathcal{L}(c') = u_1'\frac{d\langle c\rangle}{dx_1} - \frac{d\langle u_1' c'\rangle}{dx_1} \tag{8}$$

where

$$\mathcal{L} \equiv \mathcal{D}\nabla^2 - \boldsymbol{u},\nabla - \frac{\partial}{\partial t} \tag{9}$$

L is a linear stochastic differential operator containing the instantaneous velocity field $\boldsymbol{u}(x,t)$.

Equation 8 can be solved formally by using a Green's function technique (Yao et al., 1981). The result is

$$c'(x,t) = -\int_{-\infty}^{t} d\hat{t}\int_{\hat{\Omega}} d\hat{\Omega}H(\hat{t}-t+\tau_M)G(x,t\,|\,\hat{x},\hat{t})f(\hat{x},\hat{t}) \tag{10}$$

$$f(\hat{x},\hat{t}) \equiv u_1'(\hat{x},\hat{t})\frac{d\langle c\rangle}{d\hat{x}_1} - \frac{d\langle u_1' c'\rangle}{d\hat{x}_1}$$

where τ_M is a random memory time related to the bursting phenomenon observed in the near wall region and $H(\cdot)$ is a unit step function which is zero for $\hat{t} < t - \tau_M$.

An important feature of Eq. 10 is that $\tau_M < \infty$. Recent experimental observations of turbulent fluctuations in the near wall region support this hypothesis. Following Nakagawa and Nezu (1978), MacLeod and Ponton (1977) and vanDongen et al. (1978), we assume that the bursting process occurs in two dynamic stages, *viz.*, a sudden burst followed by a quick relaxation to a relatively quiescent, but still turbulent, statistically stationary state. The duration of the second stage is long compared to the first and we further assume that most of the mass transfer occurs during this less intense stage. The mean period of the second stage is $\langle\tau_M\rangle$, which controls the memory of all statistical correlations. This physical picture is also the basis for renewal theories of mass transfer (see, esp., Sideman and Pinczewski, 1975).

Because the temporal integration in Eq. 10 is limited by τ_M, the three-dimensional spatial integration will simplify considerably if $G(x,t\,|\,\hat{x},\hat{t})$ has not changed significantly from its initial spatial delta distribution during this time interval. Very near a rigid interface the effect of turbulent mixing on $G(x,t\,|\,\hat{x},\hat{t})$, although still an important factor, becomes comparable to the effect of molecular diffusion. Thus, the spatial domain over which the Green's function varies during the time τ_M is $\delta_G \equiv (D\tau_M)^{1/2}$. If τ_M is bounded, then δ_G becomes small as $D \to 0$. Note, however, that the spatial variation of $\langle c\rangle(x_1)$ and $\langle u_1' c'\rangle(x_1)$ at high Schmidt numbers also occurs over a small domain $\delta_c\ (\equiv D/\langle k_c\rangle)$, as indicated in Figure 1 and by Eq. 7. Therefore, if the D-dependence of $\langle k_c\rangle$ is stronger than ½-power, $\delta_G \ll \delta_c$ for $Sc \to \infty$. This observation suggests that a *smoothing hypothesis* applied to the average of Eq. 10 could lead to a good approximate model for the mass transfer coefficient (cf., Petty, 1975).

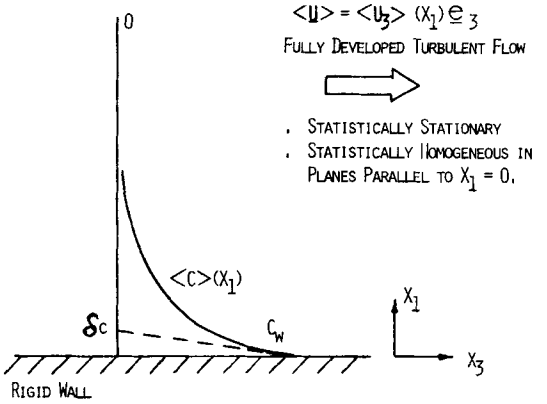

Figure 1. Fully developed mass transfer near a rigid interface.

Because $\langle c'(x,t)\rangle = 0$, Eq. 10 implies that

$$\int_0^\infty d\hat{\tau}\int_{\hat{\Omega}} d\hat{\Omega}\langle H(\tau_M-\hat{\tau})G(x,t\,|\,\hat{x},\hat{t})u_1'(\hat{x},\hat{t})\rangle\frac{d\langle c\rangle}{d\hat{x}_1} = \int_0^\infty d\hat{\tau}\int_{\hat{\Omega}} d\hat{\Omega}\langle H(\tau_M-\hat{\tau})G(x,t\,|\,\hat{x},\hat{t})\rangle\frac{d\langle u_1' c'\rangle}{d\hat{x}_1} \tag{11}$$

where $\hat{\tau} \equiv t - \hat{t}$. If the two kernels in Eq. 11 are sharply peaked about $x = \hat{x}$ for $0 \le \hat{\tau} \le \tau_M$, then Eq. 2 follows with

$$u_c(x_1) \equiv \frac{-\int_0^\infty d\hat{\tau}\int d\hat{\Omega}\langle H(\tau_M-\hat{\tau})G(x,t\,|\,\hat{x},\hat{t})u_1'(\hat{x},\hat{t})\rangle}{\int_0^\infty d\hat{\tau}\int d\hat{\Omega}\langle H(\tau_M-\hat{\tau})G(x,t\,|\,\hat{x},\hat{t})\rangle} \tag{12}$$

MODEL FOR $u_c(x_1)$

By neglecting fluctuations in the random variable τ_M, Eq. 12 simplifies to $u_c(x_1) = L(x_1)/T(x_1)$ where

$$L(x_1) = -\int_0^{\langle\tau_M\rangle} d\hat{\tau}\int d\hat{\Omega}\langle G(x,t\,|\,\hat{x},\hat{t})u_1'(\hat{x},\hat{t})\rangle \tag{13}$$

and

$$T(x_1) = \int_0^{\langle\tau_M\rangle} d\hat{\tau}\int d\hat{\Omega}\langle G(x,t\,|\,\hat{x},\hat{t})\rangle \tag{14}$$

The two space-time kernels $\langle G'u_1'\rangle$ and $\langle G\rangle$ can be derived from

$$\mathcal{L}(G) = -\delta(x-\hat{x})\delta(t-\hat{t}) \tag{15}$$

An equation for G', the fluctuating component of the Green's function, follows by subtracting the average of Eq. 15 from Eq. 15. If the nonlinear fluctuating terms are neglected (Yao et al., 1981), the resulting equation for G' can be solved using a non-random Green's function associated with the differential operator (cf. Eq. 9)

$$\mathcal{L}^0 \equiv \mathcal{D}\nabla^2 - \langle u_3\rangle(x_1)\frac{\partial}{\partial x_3} - \frac{\partial}{\partial t} \tag{16}$$

and homogeneous Dirichlet boundary conditions. Multiplying this

result for $G'(x,t|\hat{x},\hat{t})$ by $u_1'(\hat{x},\hat{t})$ and averaging yields

$$\langle G'(\cdot|\cdot)u_1'(\cdot)\rangle = -\int_0^{\langle\tau_M\rangle} d\hat{\tau} \int d\hat{\hat{\Omega}} G^0(\cdot|\cdot)\langle u_1'(\cdot)u'(\cdot)\rangle \cdot \hat{\hat{\nabla}}\langle G\rangle(\cdot|\cdot). \quad (17)$$

For high Schmidt numbers, we assume that $\langle G\rangle$ can be approximated by G^0 for short times. Furthermore, $\langle u_3\rangle(x_1)$ can also be replaced by its spatial average u_A over the concentration sublayer (see p. 467 in Monin and Yaglom, 1971). With $\langle G\rangle = G^0$, Eq. 14 implies that (see Yao et al. for a description of G^0)

$$T(x_1) = \int_0^{\langle\tau_M\rangle} d\hat{\tau}\ \mathrm{erf}\left[\frac{x_1}{\sqrt{4\mathcal{D}\hat{\tau}}}\right] = \left(\frac{4\langle\tau_M\rangle}{\pi\mathcal{D}}\right)^{1/2} x_1 + \ldots \quad (18)$$

Inserting Eq. 17 into Eq. 13 and replacing $\langle G\rangle$ with G^0 yields an expression for $L(x_1)$. For large Schmidt numbers, G^0 is sharply peaked relative to the velocity space-time correlation for $0 \le \hat{\tau} \le \tau_M$; therefore, the *smoothing hypothesis* used earlier with the gradients of $\langle c\rangle$ and $\langle u_1'c'\rangle$ can be applied again. With

$$\langle u_1'(x,\hat{t})u_1'(x,\hat{\hat{t}})\rangle = a_{11}x_1^4 \exp\left[-\frac{|\hat{t}-\hat{\hat{t}}|}{\tau_H}\right], \quad (19)$$

the resulting equation for $L(x_1)$ in the near wall region reduces to,

$$L(x_1) = \frac{h(p)a_{11}\tau_H x_1^5}{\mathcal{D}} \quad (20)$$

where $p \equiv \langle\tau_M\rangle/\tau_H$ and

$$h(p) = \frac{1}{\pi}\int_0^p \frac{d\eta}{\eta}\int_0^\eta \sqrt{\frac{\xi}{\eta-\xi}}\,e^{-\xi}d\xi = 1 - e^{-p}M\left(\frac{1}{2},1,p\right) \quad (21)$$

$M(\frac{1}{2},1,p)$ is the confluent, or degenerate, hypergeometric function (see p. 504 in Abramowitz and Stegun, 1964).

Equation 20 divided by Eq. 18 gives the following model for $u_c(x_1)$ near the mass transfer interface

$$u_c^+(x_1^+) = \alpha^+ x_1^{+4} \quad (22)$$

where

$$\alpha^+ \equiv \frac{\sqrt{\pi}}{2}\frac{h(p)}{\sqrt{p}}a_{11}^+\sqrt{\tau_H^+ Sc} \quad (23)$$

MODEL FOR THE STANTON NUMBER St^+

Substituting Eq. 22 into Eq. 7 and carrying out the integrations gives

$$St^+ = bSc^{-7/10} \quad (24)$$

where

$$b \equiv \frac{1}{\Gamma\left(\frac{1}{5}\right)}\left(\frac{625\pi h(p)a_{11}^+\sqrt{\tau_H^+}}{4\sqrt{p}}\right)^{1/5} \quad (25)$$

Petty (1975) derived a similar expression for St^+ using Eq. 12 with a different set of assumptions. In this earlier work, fluctuations in G were ignored (i.e., $G = G^0$), so the retarding effect of the turbulent flux was excluded. Moreover, the mean bursting period $\langle\tau_M\rangle$ was assumed to be long compared with the relaxation of the autocorrelation (i.e., $p \to \infty$). For $p < \infty$, this theory gives

$$St^+ = b^0 Sc^{-7/10} \quad (26)$$

where

$$b^0 \equiv \frac{5}{\pi}\sin\left(\frac{\pi}{5}\right)(\mathrm{erf}\sqrt{p})a_{11}^+\sqrt{\tau_H^+})^{1/5} \quad (27)$$

With $p = \infty$, Eq. 27 reduces to the result derived by Petty (1975).

It is noteworthy that both models predict the same Schmidt number dependence for St^+ as observed experimentally by Shaw and Hanratty (1977a). Moreover, the lead coefficients b and b^0 depend on the same three hydrodynamic parameters p, τ_H^+, and a_{11}^+, which can be estimated based on turbulent measurements in the viscous sublayer.

CONCLUSIONS

With $p = 1$, Eq. 22 gives $h(1) = 0.355$. Therefore,

$$b = 0.61\,(a_{11}^+\sqrt{\tau_h^+})^{1/5} \quad (28)$$

and

$$b^0 = 0.90(a_{11}^+\sqrt{\tau_H^+})^{1/5} \quad (29)$$

From Eqs. 28 and 29, we conclude that finite memory and the feedback effect of the flux in Eq. 10 is an important component of a mechanistic interpretation of mass transfer near rigid interfaces (also see Yao et al., 1981). Furthermore, with $\tau_H^+ = 100$ and $a_{11}^+ = 10^{-5}$, Eq. 24 gives

$$St^+ = 0.0967\,Sc^{-7/10}, \quad (30)$$

which compares favorably with the experimental data of Shaw and Hanratty [1977a], viz.,

$$St^+ = 0.0889\,Sc^{-0.704}. \quad (31)$$

Thus, the present theory gives a mean mass transfer coefficient which not only has the same Schmidt number dependence as the experimental data but basically agrees *quantitatively* with results of Shaw and Hanratty. This is in contrast to the earlier result of Petty (1975), which anticipated the unusual Schmidt number dependence of St^+ but failed to agree quantitatively with the data. For $\tau_H^+ = 100$ and $a_{11}^+ = 10^{-5}$, Eq. 26 gives $St^+ = 0.143\,Sc^{-7/10}$, which is about 60% higher than the experimental correlation Eq. 31.

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NOTATION

a_{11} = the first non-trivial coefficient in an expansion of $\langle u_1'^2\rangle(x_1)$ about $x_1 = 0$.
b, b^0 = defined by Eqs. 25 and 27, respectively
$c(x,t)$ = instantaneous concentration
$\langle c\rangle, c'$ = mean and fluctuating components of c, respectively
c_w = wall concentration
$\mathcal{D}$ = molecular diffusivity
$G(x,t|\hat{x},\hat{t})$ = instantaneous Green's function defined on the semiinfinite domain Ω associated with the operator $\mathcal{L}(\cdot)$ and homogeneous Dirichlet boundary conditions
$\langle G\rangle, G'$ = mean and fluctuating components of G, respectively
$h(p)$ = defined by Eq. 21, $p \equiv \langle\tau_M\rangle/\tau_H$
$H(\cdot)$ = unit step function
$\mathcal{L}(\cdot)$ = stochastic differential operator defined by Eq. 9
$L(x_1)$ = length scale defined by Eq. 13
Sc = molecular Schmidt number
$T(x_1)$ = time scale defined by Eq. 14
$u(x,t)$ = instantaneous velocity field
$\langle u\rangle, u'$ = mean and fluctuating components of u, resp.; u_1' is the fluctuating velocity normal to the mass transfer interface; $\langle u_3\rangle(x_1)$ is the mean velocity parallel to the mass transfer interface

$\langle u_1'(x,t)u_1'(x,\hat{t})\rangle$ = autocorrelation in the near wall region defined by Eq. 19
$\langle u_1'c'\rangle(x_1)$ = turbulent flux of a passive additive
$u_c(x_1)$ = characteristic velocity appearing in the turbulent model for $\langle u_1'c'\rangle$; $u_c \equiv L(\quad)/T(x_1)$
x = coordinate vector with components x_1, x_2, x_3
$+$ = superscript denoting a quantity made dimensionless using the wall parameters u^* and ν

Greek Letters

α = defined by Eq. 23
δ_c = characteristic size of the concentration layer near the mass transfer interface (Eq. 7)
ν = kinematic viscosity
τ_H = characteristic relaxation time for the velocity autocorrelation in a frame of reference moving with the average velocity
$\langle\tau_M\rangle$ = mean period between burst
Ω = semi-infinite spatial domain

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Evaluation of a Stochastic Model of Particle Dispersion in a Turbulent Round Jet

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The accumulation and correlation of data for empirical computations of turbulent particle dispersion is complicated since dispersion is influenced by both particle and turbulence properties. This difficulty can be circumvented by computing dispersion directly, using a stochastic particle dispersion model. The stochastic method requires an estimate of the mean and turbulent properties of the continuous phase. Particle trajectories are then computed using random sampling to determine the instantaneous properties of the continuous phase, similar to a random walk calculation. Mean dispersion properties are obtained by averaging over a statistically significant number of particle trajectories.

Several stochastic particle dispersion models have been proposed. Yuu et al. (1978) use a stochastic dispersion model, which employs empirical correlations of mean and turbulent properties, to analyze their measurements of particle dispersion in jets. Gosman and Ioannides (1981) describe a more comprehensive approach, predicting both flow properties (using a k-ϵ model) and dispersion. The latter procedure is attractive since k-ϵ models yield satisfactory predictions for many of the boundary layer type flows that are encountered with dispersion problems.

The objective of the present investigation was to reexamine the data of Yuu et al. (1978) for particle dispersion in air jets using a stochastic dispersion model similar to Gosman and Ioannides (1981). Shearer et al. (1979) have demonstrated that the present k-ϵ model provides good predictions of existing measurements of mean and turbulent properties within jets. In the following, the present stochastic dispersion model is also calibrated by comparison to predictions by Hinze (1975) and measurements by Snyder and Lumley (1971), for simpler flows, prior to applying the model to the jet dispersion data of Yuu et al. (1978).

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