cane," *Ind. Eng. Chem.* 34, 1526 (1942).

Reamer, H. H., B. H. Sage, and W. N. Lacey, "Phase Equilibria in Hydrocarbon Systems, Volumetric and Phase Behavior of the Methane-Propane System," *ibid.*, 42, 534 (1950).

———., "Phase Equilibria in Hydrocarbon Systems, Volumetric and Phase Behavior of the Methane-*n*-Heptane System," *Chem. Eng. Data Ser.*, 1, 29 (1956).

Rogers, B. L., and J. M. Prausnitz, "High-Pressure Vapor-Liquid Equilibria for Argon + Neopentane and Methane + Neopentane," *J. Chem. Thermodynamics*, 3, 211 (1971).

Sage, B. H., H. H. Reamer, R. H. Olds, and W. N. Lacey, "Phase Equilibria in Hydrocarbon Systems, Volumetric and Phase Behavior of Methane-*n*-Pentane System," *Ind. Eng. Chem.*, 34, 1108 (1942).

Shipman, L. M., and J. P. Kohn, "Heterogeneous Phase and Volumetric Equilibrium in the Methane-*n*-Nonane System," *J. Chem. Eng. Data*, 11, 176 (1966).

Manuscript received October 2, 1973; revision received October 19 and accepted October 23, 1973.

Hydrodynamics and Mass Transfer for Suspended Solid Particles in a Turbulent Liquid

G. A. HUGHMARK

Ethyl Corporation
Baton Rouge, Louisiana 70821

Levins and Glastonbury (1972) presented an excellent review and analysis for mass transfer to particles suspended in an agitated fluid. They used Tchen's equation from Hinze (1959) for the prediction of particle-turbulent fluid relative velocity. The solution using the Gaussian error curve for the Lagrangian correlation function is shown to be

$$(u_R)^2_{\text{ave}} = \frac{2(1-b)^2}{\pi}\int_0^\infty \frac{\omega^2}{(a_1^2+u_1^2)}\, T \exp - \frac{\omega^2 T^2}{\pi}\, d\omega \quad (1)$$

$$\omega_1 = \omega + c\sqrt{(\pi\,\omega/2)} \qquad a_1 = a + c\sqrt{(\pi\,\omega/2)}$$

$$b = 3\rho_f/(2\rho_s+\rho_f)$$

and for flow where Stokes law is applicable

$$a = \frac{36\,\mu}{(2\rho_s+\rho_f)d_p^2} \qquad c = \frac{18}{(2\rho_s+\rho_f)d_p}\sqrt{\frac{\mu\rho_f}{\pi}}$$

An analytic solution of Equation (1) is obtained by elimination of the Bassett term from Tchen's basic equation of motion which corresponds to $c = 0$. Levins and Glastonbury also report experimental data for fluid and particle velocities and the Lagrangian integral scales for a 25-cm diameter vessel with 7.5- and 10-cm diameter impellers. It was also observed that there were no significant differences between the root mean square fluctuating velocities of dense particles and the fluid which is consistent with the solution of the equation of motion of a particle in a turbulent fluid using the experimental Lagrangian scales of turbulence.

The objective of this paper is to utilize the Levins and Glastonbury work to develop a turbulent model for particle-liquid motion and mass transfer.

PARTICLE-LIQUID MOTION

Schartzberg and Treybal (1968) report mean and fluctuating velocities for several different impeller and vessel diameters that correlate as a function of $ND^2/(\overline{T}^2H)^{1/3}$ which corresponds to $(\epsilon D)^{1/3}$. The experimental data for fluctuating velocity reported by Levins and Glastonbury also are a function of the $(\epsilon D)^{1/3}$ term and are represented by

$$u_i = (\epsilon D)^{1/3}/3 \quad (2)$$

The Lagrangian integral scale data indicate that the length scale is approximately the width of the impeller and that the time scale can be approximated by

$$T = \frac{3\,D_i}{(\epsilon D)^{1/3}} \quad (3)$$

The a and c terms of Equation (1) represent Stokes law conditions. A more general equation for a is proposed by Schwartzberg and Treybal:

$$a = \frac{3\rho_f\,C_D\,u_R}{2(2\rho_s+\rho_f)d_p}$$

The c term can also be modified by replacing the drag coefficient response for Stokes law conditions with the drag coefficient. This results in the equation

$$c = \frac{3.67\,\rho_f}{(2\rho_s+\rho_f)}\sqrt{\frac{C_D\,u_R}{\pi\,d_p}}$$

which reduces to the c term of Equation (1) for $C_D = 24/N_{Re}$ corresponding to Stokes law conditions.

Zwietering (1958) presents correlations for suspension of sand and sodium chloride particles in low viscosity liquids with agitation conditions that are similar to those used by Levins and Glastonbury. If the minimum agitation conditions for particle suspension are assumed to correspond to the terminal settling velocity in the turbulent fluid, settling velocities can then be estimated for these conditions. Thus the relative velocity between the particle and fluid can be calculated from Equation (1) for minimum suspension conditions and this velocity could also be expected to represent the terminal settling velocity in the turbulent liquid. Figure 1 shows drag coefficients corresponding to settling velocities estimated in this manner from the Zwietering correlation for flat blade turbines with water and acetone as fluids and impeller diameters of 6 and 8 cm. Velocities were estimated by numerical integration of Equation (1) with Equation (3) for the Lagrangian time scale and the terms containing the drag coefficient for a and c. Drag coefficients for particles in a still fluid were used for the a and c terms. The drag coefficients shown by Figure 1 are observed to be greater than for particles in a still fluid. The coefficients are also observed to give a reasonably consistent plot with Reynolds number for Figure 1. Minimum agitation conditions for impeller to vessel diameter ratios of 0.27 to 0.52 were used in estimating settling velocities and drag coefficients. Figure 1 also shows a consistent correlation for this variable.

Levins and Glastonbury and Schwartzberg and Treybal also report observed settling velocity data for solid particles in agitated fluid conditions. Drag coefficients calculated from these data are also shown by Figure 1. Good agreement is observed between these data and the drag coefficients calculated from the minimum agitation suspension data.

SOLID-LIQUID MASS TRANSFER

Harriott (1962) proposed that a representative slip velocity between particles and turbulent fluid could be used for forced convection heat and mass transfer. Vectorial addition of the particle-fluid and terminal settling velocities should provide this representative slip velocity. Mass transfer data for particles in agitated fluids are reported by Barker and Treybal (1960), Harriott (1962), Keey and Glen (1966), Levins and Glastonbury (1972), Miller (1971), and Sykes and Gomezplata (1967) for solids in water, sucrose solution, ethanol, and methanol with $(\rho_s - \rho_f)/\rho_f$ ratios of 0.05 to 6.14. Impeller diameters are 5 to 10 cm for which the Lagrangian time scale represented by Equation (3) could be expected to apply. Heat and mass transfer data for ice and pivalic acid particles in water are reported by Brian, Hales, and Sherwood (1969) that are of particular interest because the solids are less dense than the fluid phase. Thus these data represent a wide range of physical property conditions.

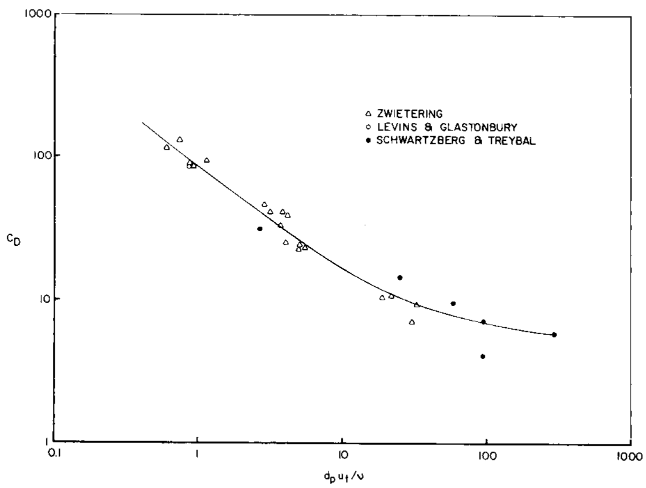

Fig. 1. Drag coefficients.

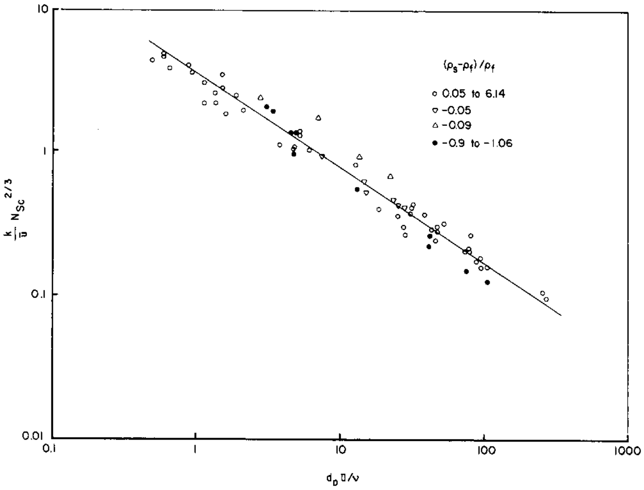

Fig. 2. Mass transfer data.

Son and Hanratty (1967) and Hughmark (1971, 1973) have presented analyses of wall region heat and mass transfer for turbulent flow in a smooth pipe. This is a developing boundary layer model of the form for mass transfer:

$$k = \alpha\, u \sqrt{f/2}\, N_{Sc}^{-2/3} \tag{4}$$

As the friction factor is a function of Reynolds number, $k\, N_{Sc}^{2/3}/u$ is also a function of the Reynolds number. The model could be expected to apply for particle heat and mass transfer in a turbulent fluid with $k\, N_{Sc}^{2/3}/\bar{u}$ as a function of the Reynolds number $d_p\, \bar{u}/\nu$. The velocity $\bar{u}$ is the vectorial sum of the terminal settling velocity in a turbulent fluid and the particle-fluid velocity:

$$\bar{u} = \sqrt{u_t^2 + u_R^2} \tag{5}$$

Figure 2 shows the mass and heat transfer data as a function of the Reynolds number with the group $k\, N_{Sc}^{2/3}/\bar{u}$ decreasing with increasing Reynolds number and the correlating equation:

$$\frac{k}{\bar{u}} N_{Sc}^{2/3} = 3.6 \left(\frac{d_p\, \bar{u}}{\nu} \right)^{-2/3}$$

or

$$\frac{k}{\bar{u}} N_{Pe}^{2/3} = 3.6 \tag{6}$$

The velocity $\bar{u}$ is obtained from Equation (5) with the settling velocity u_t calculated from Figure 1 and the particle-turbulent fluid relative velocity u_R calculated by numerical integration of equation 1 with the general equations for the a and c terms.

Data shown by Figure 2 have the restriction that the particle-fluid velocity is approximately equal to or greater than the settling velocity. Data with particle-fluid velocities less than the settling velocity show lower mass transfer coefficients than are predicted by Equation (6). The particle size range represented by Figure 2 has a limit of 80 microns for dense metal particles and 250 microns for resin particles to 0.4-cm benzoic acid particles. The upper limit for particle size would be expected to be of the order of the scale of the large eddies which is approximately the impeller width.

GAS-LIQUID MASS TRANSFER

Calderbank (1959) reports data for mass transfer from carbon dioxide gas bubbles to water and aqueous mixtures of glycol and glycerol. Bubble sizes are reported as less than 0.2 cm. Terminal settling velocities for 0.15-cm bubbles were estimated from the drag coefficients from Figure 1 and particle-fluid velocities were assumed equal to the settling velocity to obtain the representative slip velocity. These mass transfer data are also shown by Figure 2 with reasonable agreement with Equation (6).

SCALE-UP

Two scale-up methods have been proposed for particle-liquid mass transfer in agitated vessels: equal power input per unit volume and equal impeller tip speed. A number of papers apply Kolmogoroff's theory of local isotropic turbulence to particle-liquid mass transfer which implies that equal power input per unit volume is the correct scale-up method. Levins and Glastonbury (1972) discuss the application of Kolmogoroff's theory and show that experimental evidence does not support scale-up solely by power input per unit volume. Scale-up by equal impeller tip speed is indicated by the wide range of experimental conditions reported by Barker and Treybal (1960). The two procedures show much different scale-up results.

The work reported in this paper is restricted to the impeller diameter range of 5 to 10 cm as this is the region for which Lagrangian time scale data have been obtained. Lack of time scale data for larger diameter systems precludes scale-up development by this method. However, this analysis shows that mass transfer is a consistent function of Reynolds number when the energy dissipation is sufficient to provide particle-fluid velocities equal to or greater than the particle terminal settling velocity in a turbulent liquid. This implies that mass transfer scale-up should be the same as particle suspension scale-up. Zwietering's correlations show that the power input per unit volume scale-up is

$$P/V \sim D_i^{-0.55}$$

Connolly and Winter (1969) recommend equal torque per unit volume for particle suspension scale-up from data for sand suspension in 150- and 10,000-gal. vessels with downthrusting blade turbines. Thus scale-up for particle mass transfer with less than equal power per unit volume is indicated.

The analysis reported in this paper shows that mass transfer coefficients for particles with a particle-fluid velocity less than the settling velocity in a turbulent liquid do not correspond to the turbulent model for completely suspended particles. Much of the literature data appear to correspond to incomplete suspension conditions with mass transfer that is represented by a different model than for complete suspension. This apparently has clouded analysis of the particle-liquid mass transfer data.

NOTATION

a = Stokes law resistance parameter
a_1 = $a + c\sqrt{\pi\omega/2}$
b = density factor
C_D = drag coefficient
c = Basset term factor
D = impeller diameter
D_i = impeller width
D_v = molecular diffusivity
d_p = particle diameter
f = Fanning friction factor
H = liquid depth
k = mass transfer coefficient
N = impeller speed
N_{Pe} = Peclet number, $d_p \overline{u}/D_v$
N_{Re} = Reynolds number, $d_p \overline{u}/\nu$
N_{Sc} = Schmidt number, ν/D_v
T = Lagrangian integral time scale
$\overline{T}$ = vessel diameter
u = velocity
u_i = fluctuating velocity
u_R = particle-turbulent fluid velative velocity
u_t = terminal velocity
$\overline{u}$ = vectorial sum of u_R and u_t

Greek Letters

α = constant in Equation (4)
ϵ = energy dissipation rate
μ = viscosity
ν = kinematic viscosity
ρ = density
ω = frequency
ω_1 = $\omega + c\sqrt{(\pi\,\omega/2)}$

Subscripts

f = fluid
s = solid

LITERATURE CITED

Barker, J. J., and R. E. Treybal, "Mass transfer Coefficients for Solids Suspended in Agitated Liquids," *AIChE J.*, **6**, 289 (1960).

Brian, P. L. T., H. B. Hales, and T. K. Sherwood, "Transport of Heat and Mass between Liquids and Spherical Particles in an Agitated Tank," *ibid.*, **15**, 727 (1969).

Calderbank, P. H., "Physical Rate Processes in Industrial Fermentation Part II—Mass Transfer Coefficients in Gas-Liquid Contacting with and without Mechanical Agitation," *Trans. Inst. Chem. Engrs.*, **37**, 173 (1959).

Connolly, J. R., and R. L. Winter, "Approaches to Mixing Operation Scale-Up," *Chem. Eng. Progr.*, **65**, 70 (August 1970).

Harriott, Peter, "Mass Transfer to Particles: Part I. Suspended in Agitated Tanks," *AIChE J.*, **8**, 93 (1962).

Hinze, J. O., *Turbulence*, p. 352, McGraw-Hill, New York (1959).

Hughmark, G. A., "Heat and Mass Transfer for Turbulent Pipe Flow," *AIChE J.*, **17**, 902 (1971).

———., "Additional Notes on Transfer in Turbulent Pipe Flow," *ibid.*, **19**, 1054 (1973).

Keey, R. B., and J. B. Glen, "Mass Transfer from Fixed and Freely Suspended Particles in an Agitated Vessel," *ibid.*, **12**, 401 (1966).

Levins, D. M., and J. R. Glastonbury, "Particle-Liquid Hydrodynamics and Mass Transfer in a Stirred Vessel. Part I—Particle-Liquid Motion. Part II—Mass Transfer," *Trans. Inst. Chem. Engrs.*, **50**, 32 and 132 (1972).

———., "Application of Kolmogoroff's Theory to Particle-Liquid Mass Transfer in Agitated Vessels," *Chem. Eng. Sci.*, **27**, 537 (1972).

Miller, D. N., "Scale-Up of Agitated Vessels," *Ind. Eng. Chem. Process Design Develop.*, **10**, 365 (1971).

Schwartzberg, H. G., and R. E. Treybal, "Fluid and Particle Motion in Turbulent Stirred Tanks," *Ind. Eng. Chem. Fundamentals*, **7**, 1 and 6 (1968).

Son, J. E., and T. J. Hanratty, "Limiting Relation for the Eddy Diffusivity Close to a Wall," *AIChE J.*, **13**, 689 (1967).

Sykes, P., and A. Gomezplata, "Particle Liquid Mass Transfer in Stirred Tanks," *Can. J. Chem. Eng.*, **45**, 189 (1967).

Zwietering, T. N., "Suspending of Solid Particles in Liquid by Agitators," *Chem. Eng. Sci.*, **8**, 244 (1958).

Manuscript received October 12; revision received November 13 and accepted November 15, 1973.