COMMUNICATIONS TO THE EDITOR

Mass Transfer from Fixed and Freely Suspended Particles in an Agitated Vessel

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Recently Middleman (*1*) has pointed out that mass transfer relationships deduced for flows around fixed spheres may not be applied directly if the particles are freely suspended in an agitated vessel. Rather than the mean bulk flow, it is the difference between the velocity of the particle and that of the mean local flow of the entraining fluid that controls the transfer process. The object of this note is to cite some further evidence to illustrate this difference.

For the fixed-sphere experiments, a lead spherule was electroplated in copper and attached to a thin support as shown in Figure 1. This support occluded less than 2% of the surface of the sphere, which was used as the cathode in the reduction of ferricyanide ions to ferrocyanide ions in an indifferent electrolyte. This reaction has been shown to be mass transfer controlled (*2*) and the arrangement was similar to that used previously by the authors (*3*).

As in that work, a series of experiments was done with an 0.150-cm. diameter spherule held near the base and the walls of a rotating cylindrical vessel so that the local velocity was proportional to the rotational speed. Except

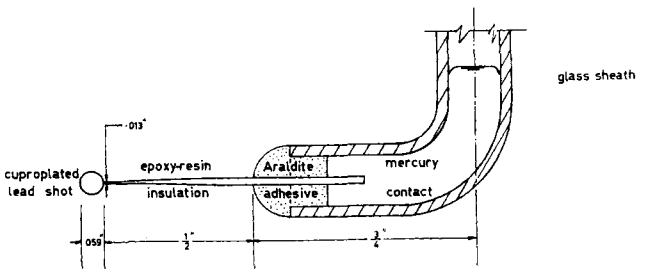

Fig. 1. Detail of spherical cathode.

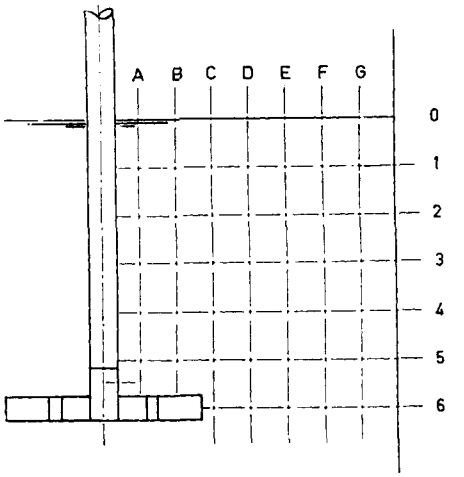

Fig. 2. Grid for locating cathode.

for a small disturbed zone, caused by the electrode and mainly downstream of it, the flow was steady at all points in the vessel. The results of twenty-six data points gave

$$k = \frac{i_L}{ZFSc_o} \sim N^{0.47} \qquad (1)$$

for

$$440 < (2\pi rN)\, d_p/\nu < 2{,}200$$

and

$$\nu/\mathcal{D} = 1{,}440$$

The correlation coefficient for the logarithmic regression was 0.99. By this relationship, mass transfer coefficients may be correlated with local velocities whenever similar

flow regimes occur. Conversely, if a dissimilar regression is found under different conditions, then the mass transfer process in that case is controlled by some other mechanism.

A second set of experiments was done with the same spherule held in a mixer in a vertical plane that bisected a segment bounded by neighbouring vertical baffles. The mixer consisted of a 7⅞ in. × 7⅞ in. diameter vessel, agitated by a centrally mounted six-bladed paddle of 2⅞ in. diameter and with a blade width/diameter ratio of ⅛. The impeller was driven through a continuously variable gearbox that gave speeds between 0 and 1,000 rev./min. The sphere was held rigidly in various positions over the plane to give a grid of locations that took in both the impeller stream and the ring vortices surrounding it (see Figure 2). To minimize disturbing the flow pattern forward of the spherule, the sphere support was placed "downstream" together with the anode and the salt bridge from the calomel cell. Again the variation of mass transfer coefficient with rotational speed was noted to yield the following results (twelve data points in each set):

Grid location	Log. regression coefficient	Correlation coefficient
C3	0.53	0.994
C5	0.44	0.993
D6	0.47	0.995
E3	0.53	0.994
F6	0.44	0.998

The difference between the regression coefficients is not statistically significant.

For the experiments with freely moving particles, *o*-nitrophenol was used. Technical grade material was purchased and steam distillated to give a solid melting at 45.0 ± 0.2°C. [reported value, 45°C. (*4*)] and of density 1.4304 g./ml. at 20°C. Molten *o*-nitrophenol was injected into a saturated aqueous solution through an eye-dropper; the droplets formed were allowed to freeze slowly during free fall. The particles were collected and the fraction between 16 and 22 B.S. mesh was kept for experiment. The same power unit was used as in the fixed-sphere experiments with six-bladed paddles for the following geometries:

Vessel diameter, in.	Impeller diameter, in.
7⅞	4
7⅞	2⅞
5½	2⅞

The continuous phase was distilled water, acidified to *p*H 3.5 ± 0.3 to suppress the dissociation of *o*-nitrophenol. The particles were introduced into the mixing vessel as quickly as possible and mixed for a set length of time, calculated from mass transfer rates taken from Barker and Treybal's correlation (*5*). The dispersed phase volume fraction was around 1.5%. At the end of each run the contents of the vessel were decanted and filtered through a fine mesh to separate the occasional chip of *o*-nitrophenol, the solution was quickly stirred to insure uniformity and sampled for ultraviolet spectroscopy. In this way, it was possible to work with concentration driving forces that differed little from the saturation concentration at the surface of each particle; yet the final mean concentration (about 7×10^{-4} g./ml.) could be detected to within ± 3%.

The results for all geometries are plotted in terms of Sherwood number $kd_T/\mathcal{D}$ against impeller Reynolds number (Nd_I^2/ν) in Figure 3. It is seen that the results fall between those of Johnson and Huang (*6*) and those of Hixson and Baum (*7*) for similar conditions at the same Schmidt number $(\nu/\mathcal{D} = 1{,}240)$. The logarithmic regression coefficient is 0.80 with a correlation coefficient of 0.943; the standard deviation of the regression coefficient is 0.10.

The results of these and the previous experiments may be summarized:

Experiment	Particle diameter, mm.	Impeller Reynolds number	Schmidt number	Regression coefficient
Held sphere	1.50	9,500 to 76,000	1,290 to 1,510	0.48 (av.)
Free sphere	0.699 to 1.00	43,000 to 125,000	1,240	0.80

These regression coefficients may be compared with values postulated from theoretical reasoning:

Experiment	Theoretical regression coefficient	Theory (*8*)
Held sphere	0.5	Laminar boundary layer with negligible wake effect
Free sphere	0.875	Boundary layer defined through skin-friction velocity in turbulent flow, $C_{DF} \sim N_{Re}^{1/4}$

Alternatively, the transport phenomena may be described in terms of particle parameters. If it is assumed that locally isotropic flow exists in the mixer, then the appropriate Reynolds number (*9*) becomes

$$N_{Re}^{*} = \rho^{2/3} (P/V)^{1/3} d_p^{4/3}/\mu \qquad (2)$$

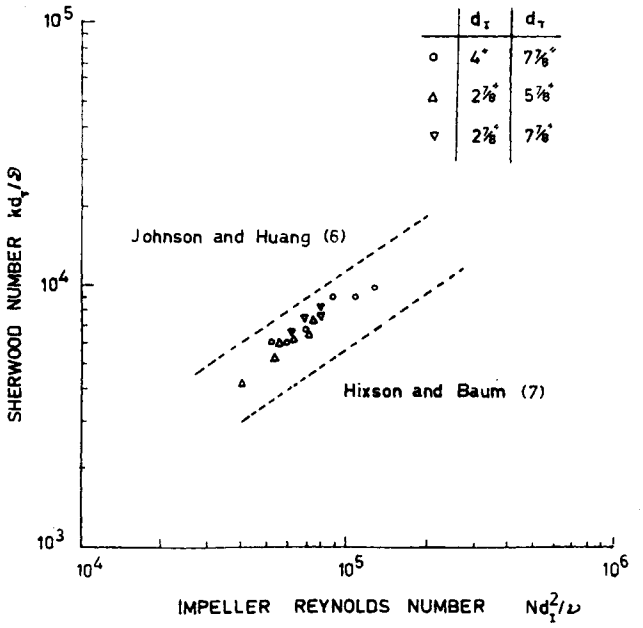

Fig. 3. Correlation of mass transfer coefficients for freely circulating particles at a Schmidt number of 1,240.

The associated Sherwood number must also be based on the mean particle diameter:

$$N_{Sh}^* = kd_p/\mathcal{D} \qquad (3)$$

The logarithmic regression of N_{Sh}^* against N_{Re}^* for free spheres yields a regression coefficient of 0.77 with a correlation coefficient of 0.916. The theoretical regression coefficient is 0.75 (9). This result lends support to the usefulness of Kolmogoroff's (10) postulate of the local isotropy of turbulent flow at high Reynolds numbers, as suggested by Middleman (1).

Therefore, the mass transfer correlations for the free spheres suggest that the transfer rate is controlled through a turbulent boundary layer, while velocity fluctuations do not disrupt the laminar boundary layer around a fixed particle. The difference between the fixed- and free-sphere experiments also clearly shows that the slip velocity that governs the mass transfer process for freely suspended particles may not be proportional to the mean bulk velocity of the fluid, as Middleman deduces (1). It would thus seem inappropriate to use correlations obtained for steady flows over fixed particles as a starting point for describing transport processes in agitated vessels. For the leaching of particles suspended in a turbulent fluid, Levich (11) obtains expressions for the mass transfer rate when the particle size is much smaller or much larger than the dissipation scale of turbulence. The predicted dependence of particle diameter on mass transfer coefficient differs from Harriott's experimental relationships (12) used by Middleman.

ACKNOWLEDGMENT

The authors wish to thank T. P. Dobbie and P. M. Farrier, who carried out the experimental work described herein. The work was supported by New Zealand University Grants Committee Grant 62/169. Also J. B. Glen acknowledges the receipt of a U.G.C. Research Fund Fellowship.

NOTATION

C_{DF} = skin friction drag coefficient
c_o = concentration at surface, g.-mole/cm.$^{-3}$
$\mathcal{D}$ = diffusivity, sq. cm./sec.$^{-1}$
d_I = diameter of impeller, cm.
d_p = diameter of particle, cm.
d_T = diameter of vessel, cm.
F = Faraday constant, (amp.)(sec.)/g.-mole^{-1}
i_L = limiting current, amp.
k = mass transfer coefficient, cm./sec.$^{-1}$
N = speed of rotation, sec.$^{-1}$
N_{Re} = d_I^2N/ν, impeller Reynolds number
N_{Re}^* = $\rho^{2/3}(P/V)^{1/3}\ d_p^{4/3}/\mu$, isotropic Reynolds number
N_{Sh} = $kd_T/\mathcal{D}$, Sherwood number
N_{Sh}^* = $kd_p/\mathcal{D}$, particle Sherwood number
P = power dissipated, (g.)(cm.)/sec.$^{-1}$
r = radial distance, cm.
S = surface area, sq. cm.
V = volume, cc.
Z = valence change
μ = viscosity, (g.)(cm.$^{-1}$)/sec.$^{-1}$
ν = kinematic viscosity, sq. cm./sec.$^{-1}$
ρ = density, g./cm.$^{-3}$

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Turbulent Motion and Mixing in a Pipe

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Lee and Brodkey (1) studied the turbulent mixing of a dye solution injected at the center of a 3-in. pipe. The observed decay of the intensity of concentration fluctuations along the centerline could be approximately predicted using Corrsin's original isotropic turbulent mixer theory (2). Since that time Corrsin has modified his theory (3). Lee and Brodkey noted in a footnote that preliminary calculations using the revised theory provided a better comparison. This note will present the fully revised computations.

The notation of reference 1 will be used, and equations presented there will not be repeated here. The modifica-