Transport of Heat and Mass Between Liquids and Spherical Particles in an Agitated Tank

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Data are reported for heat transfer from water to melting ice spheres and for mass transfer in the case of dissolving spheres of pivalic acid suspended in water agitated in a stirred vessel. The transport coefficients are found to depend on agitator power input but not on agitator design, in agreement with the Kolmogoroff theory. These experimental results are used with others in the literature to develop a correlation involving Nusselt and Prandtl or Schmidt numbers together with a dimensionless group involving agitation power. The correlation is essentially independent of solid-liquid density ratio in the range 0.8 to 1.25, and in this range the gravity group also appears to be unimportant.

Particles or drops suspended in agitated liquids are employed in various chemical processes, including crystallization, solvent extraction, polymerization, slurry catalysis, and direct-contact heat exchange. In many cases the size of the process equipment is directly related to the rate of mass or heat transfer between the particles and the liquid. Though numerous studies of the transport rates in such systems have been reported, the large number of variables involved has made it difficult to develop a full understanding of the physics of the processes of interest. Evidently the physical properties of both phases are important, as well as the nature of the turbulence in the liquid, as determined by the type of agitation and the geometry of the system.

Perhaps the most widely used method of estimating transport rates in such systems is that based on the work of Harriott (*12*). Hariott suggested that the transport rates in a stirred tank can be estimated as a multiple of the transport which would result if the particles fell through a stagnant medium at their terminal velocity. There are, however, no reliable methods of predicting this multiple.

KOLMOGOROFF'S THEORY

Several investigators (*8, 21, 27*) have suggested that Kolmogoroff's theory of isotropic turbulence may be applied to a stirred liquid in turbulent motion and, therefore, that the transport results can be correlated with the agitation power per unit volume. However, no satisfactory correlation of transport data based upon these ideas has been developed.

Kolmogoroff's theory postulates that turbulence results in a continuous inertial transfer of kinetic energy from the larger eddies to smaller and smaller eddies, finally resulting in viscous dissipation of the energy by the smallest eddies in the dissipation range. In a stirred tank, the impeller continually creates eddies described by a certain size and frequency distribution and also by a certain geometric orientation. The largest eddies are of the order of the size of the container and constitute the bulk flow. These large eddies transfer energy to smaller and smaller eddies through inertial interaction. As the kinetic energy is transferred from large to small eddies, the geometric orientation is lost. The large eddies interact to produce more random smaller ones and eventually, if sufficient interaction takes place, all directional nature is lost and the turbulent motion of the smaller eddies can be considered isotropic. These smaller eddies are in the universal equilibrium range. This range of the turbulent energy spectrum is independent of the details of the agitation device and is dependent only on the rate at which energy is supplied and the tendency of the eddies to dissipate this energy viscously. The turbulent velocities are, therefore, a function of only two parameters, the rate of energy dissipation and the kinematic viscosity of the fluid. In dimensionless form this can be written

$$\frac{vr}{\nu} = f\left(\frac{\epsilon r^4}{\nu^3}\right) \tag{1}$$

where v is the root mean square value of the relative velocity between any two points in the liquid separated by a distance r, ϵ is the turbulent energy dissipation rate per unit mass of fluid, and ν is the kinematic viscosity of the fluid. Equation (1) applies only to the turbulence associated with the small eddies in the universal equilibrium range, eddies much smaller than the impeller diameter.

If a neutrally buoyant spherical particle of diameter d is suspended in the turbulent fluid, the coefficient of mass transfer between the particle and the fluid will depend upon the diffusion coefficient, the particle diameter, the kinematic viscosity of the fluid, and the distribution of velocities in the fluid surrounding the particle. If the fluid velocities of importance are the turbulent velocities in the universal equilibrium range, a dimensional analysis yields the result:

$$N'_{Nu} = f\left(\frac{\epsilon d^4}{\nu^3}, N_{Sc}\right) \tag{2}$$

where N'_{Nu} is the mass transfer Nusselt number and N_{Sc} is the Schmidt number. In the analogous heat transfer relation these groups are replaced by the heat transfer Nusselt number and the Prandtl number, respectively.

If the particles are not neutrally buoyant in the fluid, the relative velocities between the fluid and the particle are influenced by additional variables. First, the force of gravity causes the particles to move constantly through the fluid at their terminal velocity. Therefore, Equation (2) must contain an additional group involving the acceleration of gravity, g, second, an additional relative velocity exists as the result of a difference in inertia of the two phases. An examination of a force balance on such a particle suggests that the relative motion must depend on the ratio of the densities of the two phases. Thus, if transport in a stirred tank results from locally isotropic eddies, the transport should be described by a function of the form:

$$N'_{Nu} = f\left[\frac{\epsilon d^4}{\nu^3}, N_{Sc}, \frac{gd^3(\rho_s - \rho_L)}{\nu^2 \rho_L}, \frac{\rho_s}{\rho_L}\right] \tag{3}$$

In the Kolmogoroff theory, ϵ is the rate of energy dissipation by the turbulent motion, per unit mass of fluid. In applying Equation (3) to an agitated tank, ϵ will be taken as the total agitation power per unit mass of fluid, on the assumption that essentially all of the energy is dissipated by the turbulence. This assumption should be justified when the Reynolds number of the turbulence is high, and this condition must be met if the Kolmogoroff theory is to apply.

PREVIOUS EXPERIMENTAL INVESTIGATIONS

Although the literature contains many articles which describe transport occurring in stirred tanks, the form of the function indicated by Equation (3) has not been determined, and the applicability of Kolmogoroff's theory is not widely accepted. Indeed Cutter (*9*) suggested, on the basis of indirect measurements, that the local rate of energy dissipation varies by several orders of magnitude throughout a stirred tank, and this would appear to question the validity of correlating transport data on the basis of the average agitation power. But several investigators have correlated their data as a function of the specific agitation power in order to assess the applicability of Kolmogoroff's theory, and the merit of using agitation power in correlating transport data under some circumstances has been established.

Both Oyama and Endoh (*24*), and Kneule (*18*) have been successful in correlating mass transfer data with specific agitation power for systems which varied not only in size but also in agitator geometry. Oyama and Endoh measured the rate of the dissolution of sugar in tanks having diameters of 17 and 27.4 cm., using three types of impellers. Kneule measured the dissolution rates of urea and sylvite in 3 and 50 liter tanks using several impeller-baffle combinations. Similarly, Harriott (*12*) and Kolar (*19*) correlated their mass transfer data on the basis of specific agitation power even though they did not directly make power measurements. Power input was calculated for this purpose by using the correlation of Rushton, et al. (*25*):

$$P = N_{Po} n^3 d_s^5 \rho_L \tag{4}$$

Harriott assumed the power number, N_{Po}, to be independent of the tank diameter-impeller diameter ratio. Even so, he was able to correlate within about 20% all his mass transfer data for tanks ranging in diameter from 4 to 21 in. and with six-bladed turbines varying in diameter from 1½ to 7 in. Kolar (*19*) used three different types of impellers, each requiring a different value of the power number, N_{Po}, and therefore required a separate correlation for each of his geometries.

Hixson and co-workers (*13, 15*) studied transport in stirred tanks using geometrically similar systems and later investigated the power requirements of their agitators (*14*). However, an examination of their data suggests that the particles were incompletely suspended, since the resulting transport coefficients were less than those which one might expect for the particles falling at their terminal velocity in a motionless fluid. Nevertheless, Buche (*6*) showed that these transport rates can be correlated by the specific agitation power, and Calderbank and Moo-Young (*8*) obtained reasonable success in correlating Hixson's results with transport data from fixed surfaces in a stirred tank, such as the vessel walls.

The purpose of this investigation was to test the validity of Equation (3) and to establish the form of the function, with special emphasis on the effect of the Schmidt number. All of the previous data were for mass transfer in systems with Schmidt numbers greater than about one hundred. This work has included the acquisition of data for heat transfer ($N_{Pr} = 14$), and additional mass transfer data have been obtained for comparison with the data of other investigators.

EXPERIMENTAL APPARATUS & PROCEDURE

The experimental investigations involved the measurement of heat and mass transfer rates to spheres suspended in a single stirred tank under a variety of agitation conditions. Heat transfer data were obtained by melting ice spheres in water, and mass transfer data were obtained by dissolving spherical particles of pivalic acid in water.

The experimental apparatus consisted essentially of three pieces of equipment: the apparatus to form ice spheres, that to form pivalic acid spheres, and the stirred tank in which transport rates were obtained. Spherical ice particles were formed by allowing droplets of water to fall into a column containing a mixture of toluene and octane at a temperature of − 10°C. As a result of the high interfacial tension between the two phases, spherical ice particles of about 2.4 mm. diameter were formed as the droplets solidified. The spherical particles of pivalic acid (trimethyl acetic acid, melting point 35.5°C.) were formed similarly. Drops of liquid pivalic acid were introduced into a column containing a saturated aqueous solution, and again highly spherical particles were formed as the acid solidified.

The stirred tank consisted of a 2 qt. Dewar flask. It had a diameter of 12.2 cm. and a rounded bottom such that one liter of water created a maximum depth of 10.9 cm. Two types of baffles were used. In each set four equally spaced vertical baffles extended 1.27 cm. from the vessel wall. However the larger type fit snugly against the wall, while the smaller allowed a 0.32 cm. gap between the vessel wall and the baffle. The results with the two baffle systems were the same; both gave the same power number, N_{Po}, and the same heat transfer rates.

Agitation was provided by one of two types of impellers driven by a variable speed motor. One impeller was a 6.3 cm. diam., three-bladed marine type, and the second an open turbine with four flat blades. The blade width (height) was 2.54 cm. and the overall diameter was 5.08 cm. The blade length was 2.22 cm.; that is, the blades extended to the ¼ in. shaft.

The power required to operate the impellers at speeds from 100 to 400 rev./min. was obtained experimentally by measuring the torque applied to the vessel by the agitation, by using water that contained no suspended particles. Measured values of N_{Po} at the high Reynolds numbers were approximately 0.42, 0.21, and 11.1 for the baffled marine impeller, the unbaffled marine impeller, and the baffled turbine respectively. The values of N_{Po}

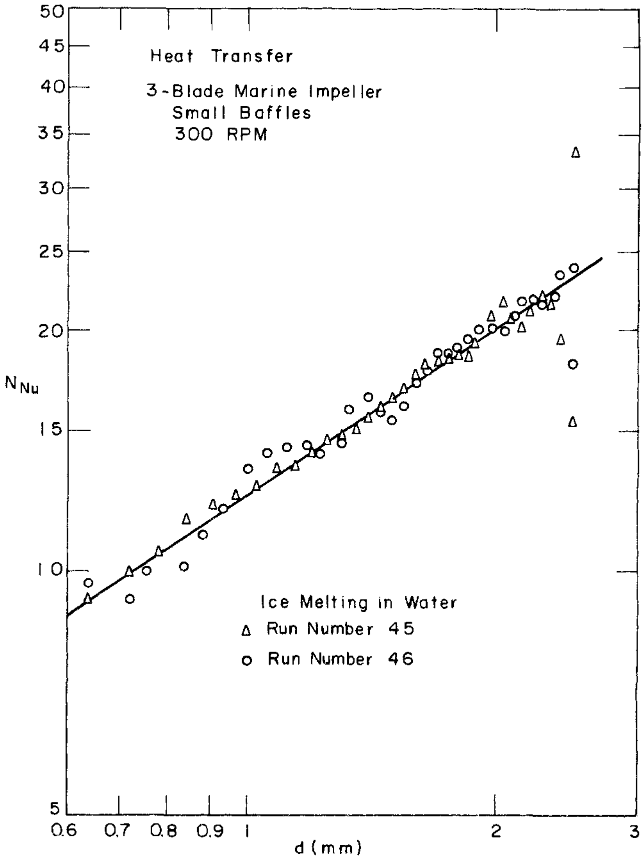

Fig. 1. Heat transfer to ice spheres.

for the marine impeller are in accord with the summary of data by Bates, et al. (*2*). The value of $N_{Po} = 11.1$ for the turbine is larger than that estimated by Equation (21) of the review by Bates, et al., which gives $N_{Po} = 8.75$. However, the data (*23*) on which the latter is based were obtained with somewhat different vessel and turbine geometries and much lower Reynolds numbers than used here.

In the heat transfer studies, measurements were made of the rate of melting of ice spheres suspended in water. Three to nine grams of uniform ice spheres (200 to 600 spheres, approximate diameter 2.4 mm.) were suddenly introduced into the agitated adiabatic tank containing water at a temperature of 0.5 to 2°C. A platinum resistance thermometer provided a continuous recording of the transient bulk water temperature as the ice spheres melted and cooled the water. This temperature transient was then used to calculate the heat transfer coefficient and the Nusselt number for the particles as a function of their diameter as they melted.

In the mass transfer studies, measurements were made of the rate of solution of pivalic acid spheres suspended in water. The spheres were suddenly introduced into the agitated tank containing water at 5°C., and then eleven samples of the bulk solution were taken at intervals of 50 to 200 sec. The acid concentration of these samples was determined by titrating each with 0.01 N sodium hydroxide in deionized water (carbon dioxide free). The resulting concentration-time data were used to calculate the mass transfer coefficient as a function of particle diameter as the particles dissolved. Concentration potential was based on the assumption that the solution adjacent to the solid surface was saturated with the acid.

The details of these experiments and the calculations are reported elsewhere (*11*).

RESULTS AND DISCUSSION

Figure 1 shows typical results of the heat transfer experiments, and Figure 2 shows the mass transfer data. All of the data of both figures were obtained under identical agitation conditions. The reproducibility of the mass transfer results is only fair, but the precision is much better for the heat transfer results. Nevertheless, a sufficient number of data points are available to determine the mass transfer coefficient with acceptable confidence, particularly at the initial diameter.

Figure 3 presents the results of this investigation plotted vs. the specific power group of Equation (3). The values of the Prandtl and Schmidt numbers in the two sets of data are $N_{Pr} = 13.7$ and $N_{Sc} = 3{,}200$. The points shown in Figure 3 represent the smooth curves through the data of several runs such as those shown in Figures 1 and 2.

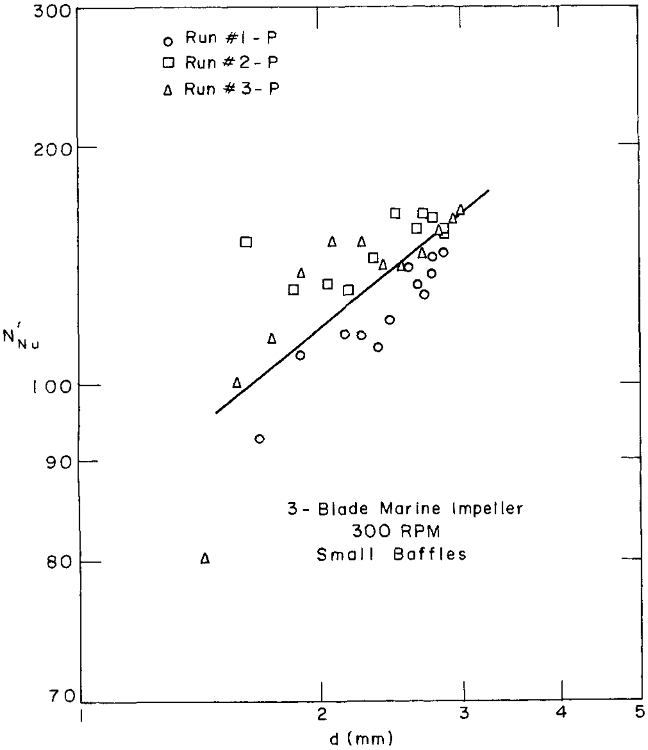

Fig. 2. Mass transfer from pivalic acid spheres dissolving in water.

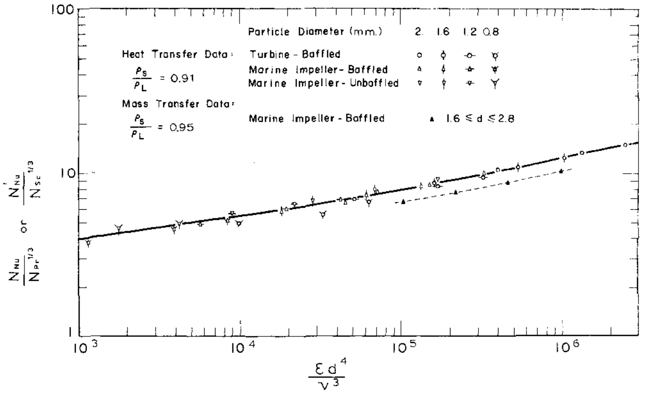

Fig. 3. Effect of dimensionless group involving specific agitation power.

The results in Figure 3 support Kolmogoroff's theory in that transport data for diverse agitation conditions are well correlated by the specific power group. The correlation brings together all of the results for heat transfer to melting ice spheres, for particle diameters from 0.8 to 2 mm. and for a hundredfold variation in agitation power. At a given agitator rev./min. the specific power varied fiftyfold from the baffled turbine to the unbaffled impeller, and the Nusselt number for a given particle size varied twofold. Thus the correlation of the heat transfer results with a single curve represents additional confirmation of the idea that specific agitation power is adequate to account for the effects of both agitator geometry and agitator speed.

This result appears not to support the inference by Cutter (9) that the local turbulent energy dissipation rate varies more than a hundredfold with position throughout a typical stirred tank. But the results in Figure 3 show that the transport coefficient varies with agitation power to only the 1/6 power, and it seems possible that the average transport coefficient might correlate well with the average energy dissipation rate even though the local dissipation rate varies widely. Furthermore, Cutter's conclusion is suspect because his evidence was quite indirect. It would appear to be quite worthwhile to obtain turbulence measurements in a stirred tank right down to the dissipation eddies in order to clarify this issue.

Figure 3 shows that significant differences exist between the mass transfer results for the pivalic acid-water system and the heat transfer results for the ice-water system. This conclusion might be questioned in view of the precision of the mass transfer data (see Figure 2). However a statistical analysis of these data for sphere diameters greater than 2.5 mm. shows that the 95% confidence limits for the broken line in Figure 3 are ± 4.2% whereas the broken line deviates from the solid line by 16%. Therefore this difference is probably a real one. It is believed that this difference is not due to a lack of analogy between heat and mass transport. Instead, it is thought to reflect the effects of the other three dimensionless groups in Equation (3). The two systems differ greatly in diffusivity and to a lesser extent in particle density and liquid viscosity. Thus the deviation of the mass transfer results from the solid curve in Figure 3 probably results either from the inadequacy of the simple 1/3 exponent to describe the effect of the Schmidt or Prandtl number or from the influence of the gravity group or of the density ratio. Additional data are required to determine the group or groups responsible for this deviation.

The data of this investigation supplement the data of

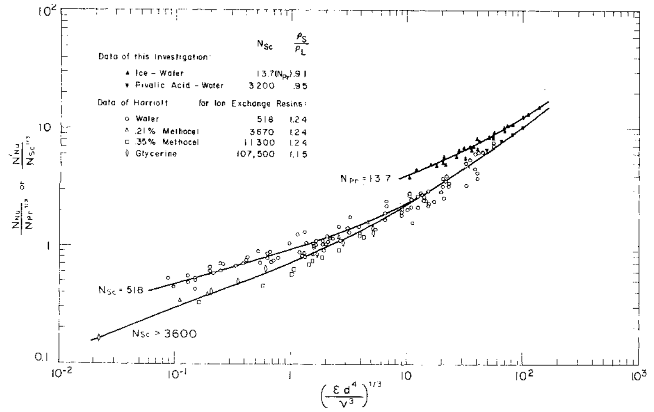

Fig. 4. Correlation of stirred tank transport data.

Harriott (*12*), and together the two sets of results can be used to determine the effects of the additional dimensionless groups of Equation (3) as well as to show the nature of function. Harriott obtained transport rates from spherical ion exchange beads suspended in a dilute sodium hydroxide solution, and although he did not directly measure the agitation power of his system, he suggests, on the basis of Rushton's (*25*) study of agitation power requirements, that the proportionality constant in Equation (4) appropriate to his studies is 6.3. By using this value, Harriott's data are compared with the data of this investigation in Figure 4.

It can be seen in Figure 4 that Harriott's mass transfer results lie below the present heat transfer data but agree with the mass transfer results of this study. Furthermore, Harriott's data for water deviate from his data for high viscosity solutions in a manner similar to that in which the present heat transfer results deviate from the mass transfer results. The 1/3 exponent is evidently inadequate to describe the effect of the Schmidt and Prandtl numbers. This inference will be tested by a detailed examination of the effects of the gravity group and the density ratio.

EFFECT OF THE GRAVITY GROUP

One can conclude from the data shown in Figure 4 that the gravity group of Equation (3) has a negligible effect on the transport rate. The arrangement of the variables in the dimensionless groups of Equation (3) includes the particle diameter in only two groups on the right-hand side, the specific power group and the gravity group. Since Figures 3 and 4 indicate that the specific power dimensionless group correlates not only the effects of the stirring speed and geometry but also the effect of particle diameter, the effect of the gravity group must be unimportant.

In order to obtain a more quantitative assessment of the effect of the gravity group, consider the data points on Figure 4 with abscissa values between 30 and 40. Harriott's data points shown in this region represent experimental conditions in which the gravity group ranges between 20.3 and 500, while the data of this investigation range in gravity group from −18.2 to −285. In effect, both sets of data contain similar broad ranges of the gravity group, since one would expect the effect of the gravity group to be independent of its sign. Over the range of data of each investigation, the points show no trend with gravity group, yet significant differences exist between the two sets of data over the same range of this parameter. Therefore, it seems clear that the gravity group of Equation (3) has negligible effect on the results shown in Figure 4.

This conclusion seems strange at first, especially when one calculates the Nusselt number, $(N_{Nu})_G$, to be expected for the particle moving through a still liquid at its terminal velocity and finds that for some of the experiments represented in Figures 3 and 4 $(N_{Nu})_G$ is as large as 50 to 70% of the measured Nusselt number in the agitated tank. The simplified analysis to follow sheds some light on this apparent contradiction, although it is by no means rigorous. The velocities causing the transport can be visualized as the result of two components: the terminal velocity resulting from the gravitational force and the turbulent velocities resulting from the liquid motion. The net velocity is the vector sum of these two components, and since the turbulent velocities exist in all directions, they may be regarded on the average as acting approximately at right angles to the terminal velocity. Thus the effective net velocity is probably nearly equal to the square root of the sum of the squares of the two components. By assuming the Nusselt number to be proportional to the square root of the velocity, as in steady flow transport from spheres (*11*), one can write

$$N_{Nu}{}^4 = (N_{Nu})_G{}^4 + (N_{Nu})_T{}^4 \qquad (5)$$

where N_{Nu} is the actual Nusselt number, $(N_{Nu})_T$ is the Nusselt number that would result from the turbulent velocities without the terminal velocity, and $(N_{Nu})_G$ is the Nusselt number which would exist if the particles moved at their terminal velocity in the absence of the turbulent velocities.

Equation (5) provides a method of estimating the effect of the gravity group for any transport data. In both Harriott's data and the present data, the values of $(N_{Nu})_G$ are always less than 70% of the measured Nusselt numbers, and therefore Equation (5) predicts that the gravity group influences the total transport by less than seven percent.

EFFECT OF THE DENSITY RATIO

Consideration of the effect of the density ratio of Equation (3) indicates that this group also has little effect upon the results shown in Figure 4. A liquid-solid density difference can be thought to cause a relative velocity between the two phases as a result of the difference in inertia. This relative velocity would be expected to increase with an increase in the difference of the densities. Hence Harriott's particles, whose density exceeds that of water by 0.24 g./cc., would be expected to undergo greater relative motion in the turbulent fluid than the ice particles used in this investigation, which differ in density from water by only 0.09 g./cc. Therefore, if the differences between Harriott's data and the present data shown in Figure 4 were the result of variations in the density ratio, Harriott's data would be expected to lie above the present data, not below. Furthermore, the present pivalic acid data agree with Harriott's data even though the density difference is quite small for the former. Finally, Harriott's data for glycerine and for methocel solutions agree with each other, even though significant differences in the density ratio exist. Therefore, it seems likely that the effect of density ratio on the data in Figure 4 is negligible, and it can be concluded that the various curves required to correlate the data are not the result of variations in this group.

Tchen (*28*) has presented a theoretical analysis of the motion of small particles suspended in a turbulent fluid. From his results, which assume that the particles are much smaller than the eddies producing the motion, the following inequality can be deduced.

$$\frac{\overline{(v_s - v_L)^2}}{\overline{v_L^2}} \leq \left(\frac{\rho_s - \rho_L}{\rho_s + \rho_{L/2}}\right)^2 \qquad (6)$$

Hence, if the root mean square turbulent velocity is known, one can obtain an upper bound on the root mean square relative velocity between the particle and the fluid. If $\sqrt{\overline{v_L^2}}$ were 10% of the impeller tip velocity, the Reynolds number based on the root mean square relative velocity between the fluid and the particle could be written

$$N_{Re} \leqq \left(\frac{0.1\pi n d_s d}{\nu} \right) \left(\frac{|\rho_s - \rho_L|}{\rho_s + \rho_{L/2}} \right) \qquad (7)$$

where n is the stirring speed and d_s is the impeller diameter. Use of Equation (7) and the assumption that the effect of this Reynolds number increase upon the measured Nusselt number is the same as for steady flow past a sphere leads to the conclusion (*11*) that the density ratio affects the data in Figure 4 by less than 12%. Furthermore, the value of 12% must be high because it corresponds to the equality rather than the inequality of Equation (7). The equality sign is valid only at high turbulent frequencies; at lower frequencies Tchen's analysis shows that smaller velocity differences result. But the magnitudes of the high-frequency turbulent velocity fluctuations are doubtless much smaller than the root mean square value, which was assumed to be 10% of the impeller tip speed. Thus it appears that Tchen's theoretical analysis supports the idea that the solid-liquid density ratio has little effect on the data in Figure 4.

For the experiments represented in Figure 4, it would appear that the dominant mechanism for producing the liquid to particle relative velocities which affect transport rates is not the inertial lag between the particle and the oscillations of large eddies, as analyzed by Tchen. Rather it appears to be the relative motion of the liquid on opposite sides of the particle which must be produced by eddies of the order of the particle size or smaller.

EFFECT OF SCHMIDT AND PRANDTL NUMBERS

Since the gravity group and the density ratio seem to have little effect on the data, the displacement of the lines on Figure 4 would appear to be due primarily to the large variation in N_{Sc} for the several systems, not adequately accounted for by employing the exponent of 1/3. It is evident by inspection that the variation with N_{Sc} is less than suggested by the $N_{Sc}{}^{1/3}$ group; if a power-law relation is appropriate the exponent must be less than 1/3.

Many other investigators (*26*) have reported the decreasing influence of N_{Sc} on N'_{Nu} as N_{Sc} increases. Elrod (*10*) has shown that if the eddy diffusivity is proportional to a power of the distance from the phase boundary, then the exponent must be 3 or greater. From this it follows that N'_{Nu} must be proportional to N_{Sc} raised to a power 1/3 or smaller, in qualitative agreement with the trend suggested by Figure 4.

OTHER EXPERIMENTAL INVESTIGATIONS

Figure 4 is based upon the data of this study and on Harriott's ion exchange data. Other experimental data will be compared with the correlation in this section.

Humphrey and Van Ness (*16*), Mattern, et al. (*20*), Kafarov and Podima (*17*), and Kolar (*19*) have reported mass transfer coefficients for solids dissolving in agitated tanks. The data of these investigators are not considered here since the calculation of mass transfer coefficients from the experimental measurements could not be carried out without assuming the manner in which the coefficient varied with particle size.

The present authors, in a companion study of ice spheres melting in salt solutions, have reported (*5*) mass transfer coefficients which lie about 35 percent below the correlation of Figure 4. This difference is thought to be the result of transient effects produced by the changing particle diameter and the transpiration at the particle surface that occurs as the ice melts. A theoretical analysis of the effects of transpiration and changing particle diameter upon the transport coefficient is presented elsewhere (*5*). This analysis indicates that these effects were negligible in all the experiments represented in Figure 4. When the theoretical corrections are applied to the ice-brine mass transfer data (*5*) the corrected results show excellent agreement with the correlation of Figure 4.

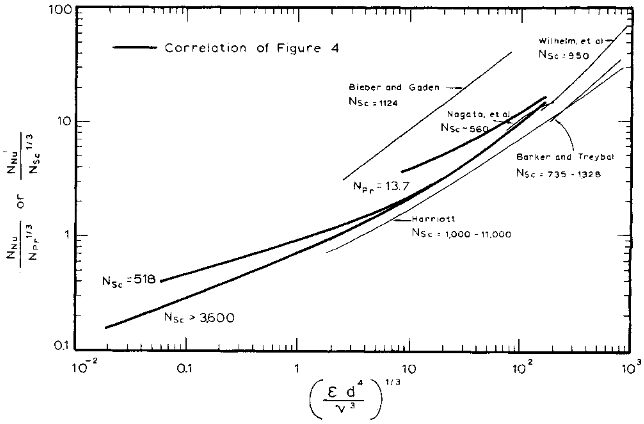

Fig. 5. Results of other investigators.

In Figure 5, results of several other investigations are compared with the correlation of Figure 4, which is shown as the heavy curves. The additional data of Harriott (*12*) are for the dissolution in water of boric, benzoic, and butyl benzoic acid particles. The data of Barker and Treybal (*1*) are for the dissolution of boric and of benzoic acid particles in water. These results agree well with each other but lie about 25% below the correlation of Figure 4. The effects of transpiration and shrinking were negligible for these data because the low solubilities of these acids result in low rates of dissolution. Furthermore, the effects of the gravity group and the density ratio seem not to have been important. A possible explanation is that kinetic effects were important and thus the dissolution rate was not mass transfer-controlled. This explanation seems unlikely because there is no tendency of the discrepancy to increase with k, as would be expected if kinetic limitations were involved. Furthermore the data of Bennett and Lewis (*3*) at comparable values of k for the dissolution of benzoic acid into water from a rotating cylinder correlated with his data on rates of solution of several metals in mercury, suggesting that the rates are wholly mass transfer-controlled. No adequate explanation has been offered for the discrepancy between Harriott's two sets of data. But it seems clear that the discrepancy is not due to an inadequacy of the specific power dimensionless group because the two sets of data were taken in the same stirred tanks and included particles of comparable size. Furthermore, the density ratio and the gravity group for the benzoic acid and boric acid particles were slightly higher than for the ion exchange particles, but those for the butyl-benzoic acid were somewhat less. Thus the discrepancy could not be explained by these changes.

Figure 5 also shows the data of Wilhelm, Conklin, and Sauer, (*30*) and of Nagata, Yamaguchi, Yabuta, and Harada (*22*) who measured transport rates by dissolving sodium chloride in water. As a result of the relatively high solubility of salt, however, these data appear to have been affected by shrinking and transpiration. The theoretical results of Brian, et al. (*5*) suggest that these effects resulted in a net reduction in the transport rate of about 23%. Therefore the lines in Figure 5 were obtained by dividing the original data by 0.77 to correct for transpiration and shrinking. Thus corrected, the results of Nagata, et al. and of Wilhelm, et al. are seen to agree well with the correlation of Figure 4.

The sugar dissolution data of Oyama and Endoh (*24*), which were correlated with a dimensionless group involving agitation power, are not shown in Figure 5 because the particle surface area was not reported and thus comparable Nusselt numbers cannot be calculated. Furthermore, the theoretical results of (5) indicate that the transport rates were greatly affected by transpiration and shrinking.

The Nusselt numbers of Bieber and Gaden (4) shown in Figure 5 are significantly greater than those of the other investigators. These results are perplexing since Bieber and Gaden's work is very similar to that of Harriott. They measured the absorption rate of copper ions on the same type of ion exchange resin as that used by Harriott. Therefore, this discrepancy cannot be the result of variations in the density ratio, and since their particles were of comparable size, the results cannot be due to the gravity group either. Bieber and Gaden suggested that the unusually large rates may have resulted from a larger effective surface area than that measured visually, but the meaning of this is not clear.

EFFECT OF PARTICLE DIAMETER

Figures 4 and 5 show that at high values of the abscissa and at high Schmidt or Prandtl number the slope of the curve is approximately ¾. Hence in this range the transport coefficient is independent of the particle diameter, as a number of investigators have found. But at low values of the abscissa in Figures 4 and 5 corresponding to small values of the specific agitation power and small particles, and at low Schmidt or Prandtl number, the slope is less than ¾, and the transport coefficient evidently varies with particle diameter. For example, the curve for $N_{Sc} = 518$ has a slope of approximately 0.3 at low values of the abscissa, whence k varies as the -0.6 power of the particle diameter (*12*). Correlations such as that presented by Calderbank (7), which show k to be independent of d, would appear to have limited applicability.

RANGE OF VALIDITY OF THE CORRELATION

The correlation of Figure 4 is believed to be valid over the range of variables indicated on the figure and to be essentially independent of density ratio as long as the solid and liquid densities are within 25% of each other. For values of the gravity group smaller in magnitude than 2,500, the effect of this group is also believed to be negligible.

The dissipation scale, η, of the turbulence is proportional to $(\nu^3/\epsilon)^{1/4}$, and Townsend (*29*) gives the proportionality constant as ½:

$$\eta = \frac{1}{2}\left(\frac{\nu^3}{\epsilon}\right)^{1/4} \tag{8}$$

Most of the viscous dissipation occurs as the result of eddies smaller in size than η, and the maximum dissipation rate occurs with eddies of a size 0.4η. By using Equation (8), the abscissa in Figures 4 and 5 can be seen to be $(d/2\eta)^{4/3}$. Therefore, the data in Figure 4 with abscissa values less than 0.4 correspond to sphere diameters less than η, the dissipation scale of the turbulence. Thus, the correlation is seen to be valid for d both greater and less than η.

On the other hand, the correlation would be expected to break down if the particle diameter were of the order of the large energy-containing eddies which are influenced by the details of the agitator geometry. Thus the correlation is evidently limited to particle diameters much smaller than the impeller diameter.

Finally, it should be emphasized that the correlation of Figure 4 is for particles completely suspended by the agitation.

CONCLUSION

The use of the specific agitation power dimensionless group results in a correlation of heat and mass transfer coefficients for spheres suspended in an agitated liquid which is independent of tank geometry, agitator geometry, and agitator speed. The functional relationship has been established for Schmidt and Prandtl numbers from 13.7 to 107,000; it is essentially independent of solid-liquid density ratio in the range 0.8 to 1.25 and of the gravity group in the range $-2{,}500$ to 2,500.

ACKNOWLEDGMENT

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NOTATION

C_p = specific heat of liquid, cal./(g.)(°C.)
D_h or D_m = molecular diffusivity for heat or mass, sq.cm./sec.
d = diameter of spherical particles, cm.
d_s = diameter of impeller, cm.
g = acceleration due to gravity, cm./sq.sec.
h = heat transfer coefficient, cal./(sec.)(cm.)(°C.)
k = mass transfer coefficient, cm./sec.
N_{Nu} = heat transfer Nusselt number, $hd/\rho_L C_p D_h$
N'_{Nu} = mass transfer Nusselt number, kd/D_m
N_{Po} = power number, defined by Equation (4)
N_{Pr} = Prandtl number, ν/D_h
N_{Re} = Reynolds number based on rms. relative velocity between fluid and particle, $d\sqrt{\overline{(v_s - v_L)^2}}/\nu$
N_{Sc} = Schmidt number, ν/D_m
n = agitator speed, rev./sec.
P = power input to agitator, erg./sec.
r = distance between two points in turbulent liquid, cm.
v = velocity, cm./sec.
ϵ = agitation power per unit mass of fluid, sq.cm./cu.sec.
η = dissipation scale of the turbulence, Equation (8), cm.
ν = kinematic viscosity of the fluid, sq.cm./sec.
ρ = density, g./cc.

Subscripts

G = resulting from terminal velocity in absence of turbulence
L = liquid
S = solid
T = resulting from turbulence in absence of terminal velocity

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A Method for Measuring Particle Diffusivity in Two-Phase Flow in the Core of a Duct

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A relatively simple, reliable means is described for measuring particle diffusivity in a particle-fluid system in the core of a duct. Particles are injected into the midstream of the duct from a line source positioned normal to the direction of fluid flow. Particle diffusivity is calculated from measurements of the standard deviations of the particle concentration profiles at various downstream distances from the injector. At an air velocity of 10 ft./sec. under room conditions, 1μ oil droplets are found to exhibit a diffusivity of about half that of air alone under the same conditions.

Whenever the diffusion equation is used to describe the behavior of the particle phase of a system in two-phase flow, knowledge of the particle diffusivity, as opposed to the fluid diffusivity, is required. Recent attempts to apply diffusion theory to electrostatic precipitation (*1*, *2*) for example, have raised serious doubts regarding the legitimacy of employing the known eddy diffusivities for fluids as reasonable approximations to particle diffusivities, even when the particles in question are of 0.9 specific gravity and as small as 1μ in mean size.

Considerable effort has been expended in determining the diffusivity of particles small enough to be sensibly affected by Brownian motion (*3*). However, comparatively little diffusivity data are available in the case of those larger particles for which eddy turbulence provides the dominant diffusive mechanism. The work of Soo, Ihrig, and el Kouh, in this connection, is noteworthy (*4*). By using stroboscopic illumination, these authors observed the paths of glass microbeads suspended in a turbulent air stream, and on this basis calculated the standard deviation, σ, of the average transverse particle displacement as a function of time, t. The particle diffusivity, D, was then found for large t, from the relation

$$D = \frac{1}{2} \frac{d}{dt} \sigma^2 \qquad (1)$$

The foregoing technique yields useful results. It is, however, subject to the disadvantage of instrumentational complexity, requiring an audio oscillator, electronic counter, multiple timer, camera, photodensitometer, and other equipment.

DIFFUSIVITY BY CONCENTRATION-PROFILE ANALYSIS

Experimental Method

The much simpler method presented here for measuring particle diffusivity is modeled after the wellknown technique of observing the rate of dispersion of a tracer fluid to determine the eddy diffusivity of its carrier fluid (*4*). In the present modification, particles whose diffusivity is to be established are introduced isokinetically through a tube or slit into the core of the duct in question. Particle-concentration profiles are then isokinetically measured by traversing the duct cross section at various downstream distances from the particle injector. Assuming turbulence that is small-scale and homogeneous, and particles of reasonable uniformity, normal-distribution profiles result, whence particle diffusivity is directly calculable from Equation (1).

The experimental arrangement used in this laboratory is shown schematically in Figure 1. Oil mist having particles of log mean diameter 1.0μ and log standard deviation 2.1 is generated by means of nozzle atomization. An automatic traversing sampler probe feeds a light-scattering

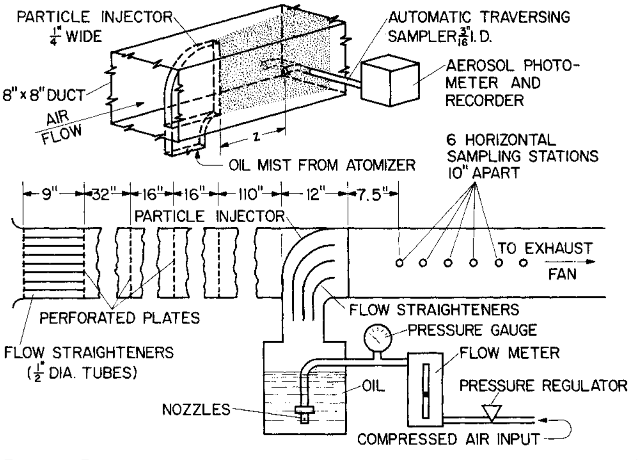

Fig. 1. Experimental duct with oil-mist generating, injecting, and sampling equipment.

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