

negative in a region larger than that defined by the separatrices shown there.

In Figure 1 the cross-hatched region is bounded by the Liapunov function tangent to the separatrix. This is the region of asymptotic stability reported in reference 1. The unshaded region is bounded by the smallest Liapunov function touching a contour of zero \dot{V} . The latter region is considerably larger than the former. Note that a portion of this extended region of asymptotic stability lies above the separatrix given in reference 1.

The extent of the stable range could be of great interest; it could be used as a measure of merit in control system design.

KRASOVSKII'S THEOREM

Krasovskii's method to find a Liapunov function has more scope than re-

ported in references 1 or 3. Briefly, the theorem as given in references 4 or 5 is as follows.

Given the following system having an isolated solution at the origin

$$\dot{x} = f(x), \text{ where } x \text{ and } f \text{ are vectors} \quad (1)$$

Define the Jacobian matrix

$$J = (J_{ij}) = \left(\frac{\partial f_i}{\partial x_j} \right) \quad (2)$$

Define a positive definite constant matrix $A = A^T$, where A^T is the transpose of A .

Then

$$f(x)^T A f(x) = V \quad (4)$$

is a Liapunov function proving asymptotic stability of (1) if

$$J^T A + A J < -Q \quad (5)$$

where Q is a positive definite symmetric matrix. [Note that (5) requires

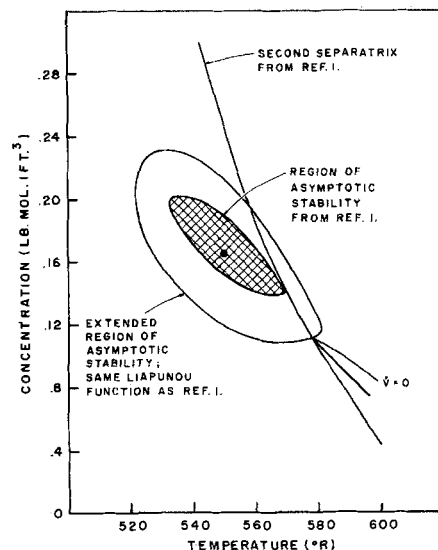


Fig. 1. Region of asymptotic stability for a chemical reactor.

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Mass Transfer from Particles in Agitated Systems: Application of the Kolmogoroff Theory

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Many industrially important fluid-particle transfer operations are carried out in agitated systems. An analytical description of such turbulent transport phenomena is virtually impossible. While some data exist on the detailed flow field in an agitated baffled vessel (7), no experimental study of the relative flow field experienced by a particle free to move with the fluid has been carried out. Of course, transport rates are governed by the flow relative to the particle, and hence might be different for two different particles in the same agitated vessel.

The usual approach to predicting the behavior of such a complex system is to assume, as a framework for a theory, a simple model for which an analytical or semi-empirical description exists, and to modify certain elements of the model in order to account for known experimental results. For example, in describing mass transfer by forced convection from spherical particles, one often starts with the well-known Frössling equation (4, 10)

$$N_{Sh} = 2 + 0.6 (N_{Re})^{1/2} (N_{Sc})^{1/3} \quad (1)$$

which holds for transfer from single spheres in the absence of natural convection.

A fundamental objection to the use of Equation (1) lies in the fact that the Frössling equation is established for steady state flows, whereas a particle in an agitated vessel experiences varying mean relative velocities which depend upon the path of the particle through the vessel. Hence, transient effects might be of some importance in such a process, and might lead to transport laws of a form different from that of the Frössling equation. Even if Equation (1) is applicable to a particle in an agitated system, one must still estimate an appropriate velocity to use in the Reynolds number.

In what follows it is implied that steady state correlation such as the

Frössling equation form a suitable framework for a theory applicable to agitated systems. The purpose of this note is to discuss rational methods of predicting the manner in which macroscopic agitation parameters enter such a correlation. It is proposed that the Kolmogoroff theory of universal equilibrium leads to useful results. The test of this approach is made with some data previously published by Harriott.

The Frössling equation is obtained from a boundary layer development which is most appropriate at high Reynolds numbers, when the flow relative to the sphere is turbulent. For small enough particles in an agitated tank it might happen that the Reyn-

TABLE 1. MASS TRANSFER CORRELATIONS

$kD_P/\mathcal{D} - 2 = K D_P^{2/3} \mathcal{D}^{1/3} N^{1/2} \nu^{-1/6} \mathcal{D}^{-1/3}$	$D_P \gg \eta$	(1)/(6)
$kD_P/\mathcal{D} - 2 = K D_P^{1/2} \mathcal{D}^{1/4} N^{3/8} \nu^{-1/24} \mathcal{D}^{-1/8}$	$D_P \ll \eta$	(1)/(7)
$kD_P/\mathcal{D} - 2 = K D_P \mathcal{D}^{1/2} N^{3/4} \nu^{-5/12} \mathcal{D}^{-1/3}$	$D_P \ll \eta$	(1)/(8)
$kD_P/\mathcal{D} - 2 = K D_P^{4/9} \mathcal{D}^{2/9} N^{1/3} \mathcal{D}^{-1/3}$	$D_P \gg \eta$	(2)/(6)
$kD_P/\mathcal{D} - 2 = K D_P^{1/3} \mathcal{D}^{1/6} N^{1/4} \nu^{1/12} \mathcal{D}^{-1/3}$	$D_P \ll \eta$	(2)/(7)
$kD_P/\mathcal{D} - 2 = K D_P^{2/3} \mathcal{D}^{1/3} N^{1/2} \nu^{-1/6} \mathcal{D}^{-1/3}$	$D_P \ll \eta$	(2)/(8)
$kD_P/\mathcal{D} - 2 = K D_P^{4/3} \mathcal{D}^{2/3} N \mathcal{D}^{-1}$	$D_P \gg \eta$	(3)/(6)
$kD_P/\mathcal{D} - 2 = K D_P \mathcal{D}^{1/2} N^{3/4} \nu^{1/4} \mathcal{D}^{-1}$	$D_P \ll \eta$	(3)/(7)
$kD_P/\mathcal{D} - 2 = K D_P^2 \mathcal{D} N^{3/2} \nu^{-1/2} \mathcal{D}^{-1}$	$D_P \ll \eta$	(3)/(8)

olds number for the sphere is small (say $N_{Re} < 1$), even though the gross motion in the system is obviously turbulent. Thus one might expect that a more appropriate equation for this case would be one derived for transfer from a sphere when the relative flow in the neighborhood of the sphere is laminar.

Examples of such theories are those developed by Friedlander (3) for the cases of large Peclet number

$$N_{Sh} = 0.991 (N_{Pe})^{1/3} \quad N_{Pe} > 10^2 \quad (2)$$

and small Peclet number

$$N_{Sh} = 2(1 + 0.25N_{Pe} + 0.083N_{Pe}^2 + \dots) \quad N_{Pe} < 1 \quad (3)$$

both of which are valid only at small Reynolds number ($N_{Re} < 5$). Again one faces the problem of selecting an appropriate velocity, in this case for the Peclet number, if he wishes to apply these equations to an agitated tank.

THE KOLMOGOROFF THEORY

Basically, the Kolmogoroff theory is a sophisticated form of dimensional analysis. The details of the theory are well described in the text by Hinze (6), and an excellent description of the basic ideas is given in the work by Shinnar and Church (12). Of particular interest is the postulate that there is some length scale, η , which is a characteristic of the turbulent field. It is a rough measure of the size of eddies mainly responsible for viscous dissipation of energy. Kolmogoroff further postulates that if one has a particle large in comparison with η , but small with respect to macroscopic dimensions of the turbulent system, and if this particle is free to move in the turbulent field, then the dynamics of the particle is controlled only by the particle diameter, D_p , and the rate of energy dissipation per unit mass, ϵ . In particular, macroscopic dimensions of the system, and the fluid viscosity, are assumed unimportant, except insofar as they might appear in ϵ . This regime of flow is known as the inertial subrange.

The dissipation scale is usually assumed to be of an order of magnitude given by (12)

$$\eta = \epsilon^{-1/4} \nu^{3/4} \quad (4)$$

where ν is the kinematic viscosity of the fluid. For a well baffled agitated tank it is common to take (11)

$$\epsilon = K_1 N^3 D^2 \quad (5)$$

One now postulates that if $D \gg$

TABLE 2. SUMMARY OF SYSTEMS STUDIED BY HARRIOTT

All runs at 300 r.p.m. with 2-in. turbine in 4-in. tank

Ion exchange resin spheres in:	μ , poise	N_{Sc}	$(N_{Re})_{tank}$	D_p , microns	η , microns
Water	0.01	518	12,500	10-600	45
0.2% methocel solution	0.067	3,670	1,850	40-300	180
0.35% methocel solution	0.20	11,300	625	15-600	430
Glycerine	0.18	107,500	563	15-600	430
Benzoic acid spheres in water	0.01	1,300	12,500	130-8000	45

$D_p \gg \eta$, then the mean relative velocity in the neighborhood of a particle depends only on D_p and ϵ . From dimensional considerations, the only quantity with the dimensions of velocity is

$$u = K_2 D_p^{1/3} \epsilon^{1/3} \quad (6)$$

On the other hand, if the particles are so small that $D_p \ll \eta \ll D$, then Kolmogoroff postulates that the particle dynamics may depend upon the kinematic viscosity, ν , in addition to D_p and ϵ . This regime of flow is called the viscous subrange.

Kolmogoroff assumes that D_p is unimportant in the viscous subrange, and takes

$$u = K_3 (\nu \epsilon)^{1/4} \quad (7)$$

If D_p is assumed important, then there is no unique expression for u obtainable through dimensional considerations alone, since $D_p(\epsilon/\nu^3)^{1/4}$ is dimensionless, and could appear as a factor to any power in Equation (7). One finds, for example (12)

$$u = K_3 D_p (\epsilon/\nu)^{1/2} \quad (8)$$

and (9)

$$u = K_3 D_p^2 (\epsilon^3/\nu^5)^{1/4} \quad (9)$$

appearing in the literature.

It is now assumed that the velocity \bar{u} appropriate to Equations (1), (2), and (3) is proportional to the velocity u given in Equations (6), (7), (8), and (9). If one introduces Equations (6), (7), (8), and (9) into Equations (1), (2), and (3) and uses Equation (5) for ϵ , a set of relations is obtained which can be subjected to experimental examination. The results are shown in Table 1, where the notation (1)/(6) means Equation (1) used with Equation (6).

Results using Equation (9) are not tabulated. We note that Equations (1)/(6) and (2)/(8) are identical, although they stem from different models for both N_{Sh} and \bar{u} .

COMPARISON WITH EXPERIMENT

The most convenient variable to examine as a test of these theories is the

particle size D_p . The dependence of k on D_p varies from -0.67 to $+1.0$, depending on the equation selected. Since D_p can be easily varied by three orders of magnitude, a determination of the dependence of k on D_p should yield a critical experiment.

An excellent set of such data already exists in the literature, given by Harriott (5). A summary of the systems examined is given in Table 2. His results indicate that k decreases with D_p , with a power of D_p somewhere between -1.0 for small particles and -0.33 for large particles. This observation eliminates Equation (1)/(8), and all equations based on Equation (3), as being applicable to Harriott's data.

Since the particle velocities are unknown we have no basis upon which to estimate Reynolds numbers. Thus, we cannot argue for or against the applicability of the Frössling equation as opposed to the low Reynolds number Peclet-type correlations. Hence, we shall examine the applicability of Equations (1)/(6), (1)/(7), and (2)/(6), (2)/(7), and (2)/(8) to Harriott's results.

For this purpose dimensionless groups are introduced, and upon rearrangement we find

$$\frac{N_{Sh} - 2}{(N_{Sc})^{1/2}} = K(N_{Re})^a (D_p/D)^b \quad (10)$$

Here we have introduced ND^2/ν as a "tank" Reynolds number. The exponents a and b are summarized in Table 3.

In order to test the applicability of these equations we must be able to estimate η for Harriott's data. Shinnar (12) quotes a value of η of 25 microns

TABLE 3. EXPONENTS FOR EQUATION (10)

a	b		
1/2	2/3	$D_p \gg \eta$	(1)/(6)
3/8	1/2	$D_p \ll \eta$	(1)/(7)
1	4/9	$D_p \gg \eta$	(2)/(6)
1/4	1/3	$D_p \ll \eta$	(2)/(7)
1/2	2/3	$D_p \ll \eta$	(2)/(8)

* Kolmogoroff's discussion implies a neutral density particle. One should include a dependence on the density difference ratio, $\Delta\rho/\rho$. This point will be discussed later.

for water when the energy input is 1 hp. per 100 gal. Assuming ϵ and the energy input per unit volume are proportional, then we may rewrite Equation (4) and find η (in microns) from

$$\eta = 25(\epsilon')^{-1/4} \nu^{3/4} \quad (11)$$

where ϵ' is in hp. per 100 gal. and ν is in centistokes. Values of η are shown in Table 2.

Figure 1 shows a test of Equation (10), for $a = 1/2$ and $b = 2/3$. The large and small particle data are distinguished by using open or filled symbols, respectively. Considering the wide range of variables, the correlation is seen to be fairly good. A similar test of Equation (2)/(6) yielded separate lines for each set of data, with the separation being as much as an order of magnitude in the abscissa. It would appear that a model based on Equation (2)/(6) may be rejected.

A slope of unity for the large particle data would be in agreement with Equation (1)/(6). The slope of the upper portion of the curve is found to be about 1.2. A slope of unity for the small particle data would confirm Equation (2)/(8). The slope of the lower portion of the curve is about

0.75. Equations (1)/(7) and (2)/(7) predict slopes of 0.75 and 0.67, respectively, for the lower portion of the data.

The values of η given in Table 2 are, at best, order of magnitude estimates. Thus, we are in no position to argue about where one theory is applicable compared with another, with respect to particle size. Furthermore, the small amount of data, and the scatter in these data, make it difficult to argue for a slope of 0.75 compared to a slope of 0.67. Hence, we refrain from any attempt to argue further among the individual correlations. We do observe, however, that the Frössling equation in conjunction with the Kolmogoroff theory predicts a correlation in reasonable agreement with the data at hand.

It is appropriate to remark that the application of the Kolmogoroff theory to mass transfer from particles is not a new idea. Levich (9) presents results that can be shown equivalent to Equations (1)/(8) and (3)/(6), but examines no data.

Calderbank (2) applies the theory, but fixes the exponent of the Reynolds number to agree with the "observation"

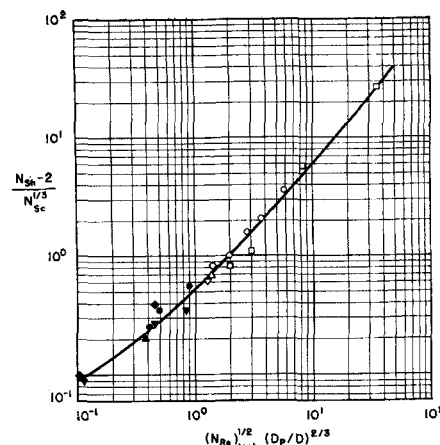


Fig. 1. Harriott's data (his Figure 8) replotted according to Equation (10), with $a = 1/2$ and $b = 2/3$. Ion exchange resin in: \circ water; \triangle 6.7 cp methocel; ∇ 20 cp methocel; \diamond 18 cp glycerine. Benzoic acid in water \square . Filled symbols represent data for $D_p < \eta$; open symbols for $D_p > \eta$.

that k is independent of D_p . The data of Barker and Treybal (1) are cited in support of this observation. Their particle sizes in a given run varied by a factor of two or three and were

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First-Order Reaction and Anisotropic Diffusion in Flowing Media

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Studies on the mass transfer in fluids moving through porous materials suggest that it may be useful to introduce anisotropic effective diffusivities, represented by a tensor quantity (1). Thus the molar flux density of a component A, referred to stationary coordinates (noncoincident with the principal axes of diffusion), should be

$$N_{Ai} = -D_{ij} \frac{\partial c_A}{\partial x_j} + c_A u_i \quad (1)$$

where i, j, \dots are dummy suffixes indicating the components of vectors and tensors along the axes, and Einstein's summation convention is adopted.

If the component A reacts with a first-order kinetics, the continuity equation, introducing expression (1), becomes

$$\frac{\partial c_A}{\partial t} = -u_i \frac{\partial c_A}{\partial x_i} + \frac{\partial}{\partial x_i} D_{ij} \frac{\partial c_A}{\partial x_j}$$

$$- \left(k + \frac{\partial u_i}{\partial x_i} \right) c_A \quad (2)$$

where, to maintain the generality, fluid density is not assumed constant.

In previous papers (2 to 4) Dankwerts' relation (5), giving the concentration profiles for unsteady diffusion in systems with first-order reaction in terms of those for nonreacting systems of the same geometry and with identical boundary conditions, is extended to flowing media with constant density and diffusivity.

Following the development given in the previous papers (2 to 5), it is easy to verify that if it is

$$\frac{\partial \bar{c}_A}{\partial t} = L \bar{c}_A \quad (3)$$

$$\bar{c}_A(0, x) = 0 \quad (4)$$

where L is an operator depending only upon position and containing only spatial derivatives, Dankwerts' relation

$$c_A = k \int_0^t \bar{c}_A(\lambda, x) e^{-k\lambda} d\lambda + \bar{c}_A e^{kt} \quad (5)$$

gives an integral of the equation

$$\frac{\partial c_A}{\partial t} = (L - k) c_A \quad (6)$$

where k is a constant. Obviously it is

$$c_A(0, x) = 0 \quad (7)$$

It is also easy to verify that if \bar{c}_A satisfies boundary conditions of the following types

$$\bar{c}_A(t, x) = c_0(x) \quad (8)$$

$$\lambda_i D_{ij} \frac{\partial \bar{c}_A}{\partial x_j} = 0 \quad (9)$$

$$-D_{ij} \frac{\partial \bar{c}_A}{\partial x_j} + \bar{c}_A u_i = f_i(x) \quad (10)$$

the same conditions are satisfied by c_A .

Thus the solution of Equation (2), using (7), (8), and (9), may be obtained

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INFORMATION RETRIEVAL

(Continued from page 758)

Water-oil displacements from porous media utilizing transient adhesion-tension alterations, Michaels, Alan S., and Marcellus C. Porter, *A.I.Ch.E. Journal*, 11, No. 4, p. 617 (July, 1965).

Key Words: A. Recovery-8, 7, Oil-9, 2, Porous Media-9, Displacement-8, 10, Water-10, 9, Reverse-Wetting Agent-6, Hexylamine-6, Amines-6, Wettability-6, Flow Rate-6, Laboratory Scale-0, Two-Phase Flow-9. B. Formation-8, Masses-9, Oil-9, Laboratory Scale-0.

Abstract: Laboratory studies have demonstrated that the injection of small quantities of reverse-wetting agents during water-displacement can increase oil recovery from unconsolidated porous media. Hexylamine is a suitable reverse-wetting agent. The mechanism by which large oil masses are formed and propagated was studied in this investigation.

Local parameters in cocurrent mercury-nitrogen flow: Parts I and II, Neal, L. G., and S. G. Bankoff, *A.I.Ch.E. Journal*, 11, No. 4, p. 624 (July, 1965).

Key Words: A. Velocity-8, Liquid-9, Void Fraction-8, Two-Phase Flow-9, Mercury-9, Nitrogen-9, Power Law-8, Slug Flow-9, Intensity-8, Density-9, 8, Mixing Length-8, Resistivity Probe-10, Impact Probe-10.

Abstract: An experimental study was made in a cocurrent vertical mercury-nitrogen flow of local parameters, including local liquid velocity and gas fraction. Power-law continuity expressions are derived. Experimental velocity and void profiles are given for the mercury-nitrogen flow. Intensities of density and liquid velocity fluctuations are computed for the first time, and a comparison is made with Levy's mixing-length theory.

Effectiveness factors in a nonisothermal reaction system, Cunningham, R. A., J. J. Carberry, and J. M. Smith, *A.I.Ch.E. Journal*, 11, No. 4, p. 636 (July, 1965).

Key Words: Hydrogen-1, Ethylene-1, Ethane-2, Coke-3, Copper-Magnesium Oxide-4, Temperature-6, Catalyst Density-6, Hydrogenation-9, Effectiveness Factor-8, Catalysis-8, Reactor-10, Activation Energy-9.

Abstract: Hydrogenation rates of ethylene in a copper-magnesium oxide catalyst were measured for fine catalyst particles and 1/2-in. spherical pellets from 60° to 160°C. Effectiveness factors for this exothermic reaction system ranged from 0.2 to 25 depending upon the temperature and density of the catalyst pellets. The activation energy for the particles was 11,800 cal./g. mole (above 120°C.), while E for the pellets decreased to zero at high temperatures. Reasons for this behavior are discussed.

The effective thermal conductivity of the catalyst pellets was measured as a function of density. The effective diffusivity was estimated from pore size and pore volume measurements.

Dynamics of bullet shaped bubbles encountered in vertical gas liquid slug flow, Street, James R., and M. Rasin Tek, *A.I.Ch.E. Journal*, 11, No. 4, p. 644 (July, 1965).

Key Words: A. Two-Phase-0, Flow-8, Gases-9, Bubbles-9, Fluids-9, Pipes-9, Vertical-0, Slug Flow-8, Mass Balance-8, Momentum Balance-8, Shape-8, 9, Holdup-8, 9, Shear Stress-8, 9, Velocity Profile-8, 9, Prediction-8, Mathematical Model-10, 8.

Abstract: A theoretical analysis is presented of the liquid film flowing around long, bullet shaped gas bubbles which characterize the slug-flow regime in two-phase flow through vertical pipe. Integral mass and momentum balances on the liquid film allow the prediction of gas bubble shape, liquid holdup around the bubble, wall shear stress acting on the liquid film, and velocity profiles in the liquid film.

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large enough that D_p would be larger than η . Such results cannot be explained on the basis of any theories presented here. Harriott offers some criticisms of the work of Barker and Treybal, but indicates that, for a certain size range, his "slip velocity" theory predicts an absence of an effect of D_p on k .

LIMITATIONS OF THE THEORY

It is important to note that the tank Reynolds numbers for these data range from about 560 to 12,500. It is generally recommended that Equation (5) not be used unless ND^2/ν exceeds 50,000 (12). However, the basis of this recommendation lies in the observation that, for sufficiently high values of ND^2/ν , the power number vs. tank Reynolds number curve is flat (11), and Equation (5) holds. Examination of the data of reference 11 indicates that this curve is nearly flat for tank Reynolds numbers beyond 600, when the ratio of baffle width to tank width is 10%, as in Harriott's work. Hence, we should expect no large error in the use of Equation (5).

More fundamental is the criticism that the Kolmogoroff theory may not hold if the flow field is not sufficiently turbulent. There does not appear to be any clear-cut method of deciding whether a flow is so turbulent that an inertial subrange exists. One criterion, of course, could be based on a demonstration that the Kolmogoroff theory describes the small scale dynamic behavior of a given system.

An important variable that has been neglected here is the density difference between particle and liquid. Two particles which differ only in density, if placed in an identical system, will experience different relative turbulent flow fields because their inertial response to the fluctuating flow will differ. Presumably, a minute particle of neutral density would experience no relative motion in its immediate neighborhood, and would lose mass strictly by diffusion ($N_{sh} = 2$).

Analyses presented by Levich indicate that the velocity u should include a factor $(\Delta\rho/\rho)^{1/2}$ for $D_p \gg \eta$ and $(\Delta\rho/\rho)$ for $D_p \ll \eta$. The data examined here do not have a sufficient variation in $\Delta\rho/\rho$ to warrant a critical test. However, since $k \sim \bar{u}^{1.2}$ at high transfer rates, one would expect k to vary with the 0.25 to 0.50 power of $\Delta\rho/\rho$. Harriott observes that k varies with the 0.3 to 0.4 power of $\Delta\rho$ for some heavy particles tested.

The success of this method of correlation must be viewed only as a

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direct confirmation of the predicted dependence of the Sherwood number on particle size and Schmidt number, since these were the major variables in the data examined. However, the predicted dependence of k on turbine speed and liquid viscosity is consistent with observations cited by Harriott.

It would appear that the Kolmogoroff theory offers a framework within which a useful correlation of data on mass transfer from particles in agitated systems may be obtained.

NOTATION

- \mathcal{D} = diffusivity, sq. cm./sec.
 D = impeller diameter, cm.
 D_p = particle diameter, cm.
 k = mass transfer coefficient, cm./sec.
 K_1 , etc. = dimensionless constants
 N = impeller speed, r.p.s.
 $N_{Pe} = D_p u / \mathcal{D}$ = Peclet number
 $N_{Re} = \bar{u} D_p / \nu$ = Reynolds number
 $(N_{Re})_{\text{tank}} = ND^2 / \nu$ = tank Reynolds number
 $N_{Sc} = \nu / \mathcal{D}$ = Schmidt number
 $N_{Sh} = k D_p / \mathcal{D}$ = Sherwood number
 \bar{u} = average velocity in neighborhood of a particle, cm./sec.

Greek Letters

- ϵ = energy dissipation rate per unit mass, sq. cm./sec.³
 η = scale of dissipating eddies, microns
 μ = viscosity, poise
 ρ = liquid density, g./cc.
 $\Delta\rho$ = density difference between particle and liquid, g./cc.
 ν = kinematic viscosity, sq. cm./sec.

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