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Mass Transfer to Drops Moving Through Power Law Fluids in the Intermediate Reynolds Number Region

The mass transfer rate to fluid spheres is calculated for power law and Newtonian fluids by using the intermediate Reynolds number stream functions of Nakano and Tien (1970) and Yamaguchi et al. (1974), respectively. The *Sh* increases with increases in *Re* and *Pe* and decreases in *n*. Better results are obtained with Nakano and Tien's functions when $Re > 10$ and with Yamaguchi's functions when $Re < 10$.

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SCOPE

Mass transfer from bubbles and drops is an important and active research area in the field of chemical engineering. Its subject matter has important applications in the design of contacting equipment for processes such as distillation, gas absorption, and liquid-liquid extraction. Large numbers of experimental and theoretical studies have been published on systems in which both phases are Newtonian fluids. However, in recent years the number of physical systems for which the fluid mechanics can only be described by non-Newtonian models has been increasing. The following are examples of industrial processes in which non-Newtonian phases are encountered: activated sludge processes, pharmaceutical production, polymerization processes, lubrication oil production, paint production, and food processing. In order to cope with the problem of estimating mass transfer rates in non-Newtonian systems, a number of researchers have recently focused their efforts on non-Newtonian dispersed phase phenomena.

Gürkan and Wellek (1976) numerically solved the diffusion equation for the creeping flow of a Newtonian droplet in a power law type of fluid by using the Mohan (1974) stream functions. Nakano and Tien (1970) have presented a set of stream functions for a drop moving through a power law type of fluid in the intermediate Reynolds number region. Experimental liquid-liquid extraction data have been presented by Schafermeyer et al. (1975) for the case of mass transfer to a droplet from a non-Newtonian continuous phase when the resistance to mass transfer is in the continuous phase. Yamaguchi et al. (1974) proposed a new set of stream functions, the use of which enabled them to predict droplet drag and continuous phase mass transfer coefficients with a smooth transition from the creeping flow regime to the intermediate Reynolds number region; their analysis involved only Newtonian fluids. However, we believe their type of stream functions may prove useful for the study of non-Newtonian fluid flow around drops. Because the Reynolds numbers encountered in droplet phenomena often fall into the range of the intermediate Reynolds number region, the prediction of the rate of mass transfer in this region for both Newtonian and non-Newtonian fluids is important and is, therefore, the subject of the current investigation.

The specific objectives of the present study are: to quantitatively predict the continuous phase Sherwood number for a Newtonian droplet moving through a power law type of continuous phase in the intermediate Reynolds number region, to determine the effect of the Reynolds number and the power law flow behavior index on the Sherwood number in the intermediate Reynolds number region, and to compare the theoretical results, which are obtained by using the Nakano and Tien (1970) and the Yamaguchi et al. (1974) types of stream functions, with available experimental data.

CONCLUSIONS AND SIGNIFICANCE

The results obtained from the solution of the mathematical model show that in the intermediate Reynolds number region, the continuous phase Sherwood number increases with a decrease in the flow behavior index n and with an increase in the Reynolds and the Peclet numbers for flow through a power law fluid. The mass transfer results obtained in this study by using the Nakano and Tien (1970) stream functions do not converge, as the Reynolds number is decreased, with the results of Gürkan and Wellek (1976) which were developed by using the

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stream functions of Mohan (1974) for the creeping flow region. This suggests that the stream functions developed by Nakano and Tien for the intermediate Reynolds number region can not be successfully extended to cover the region $Re' < 5$. The theoretical results of the present study provide an upper bound to the mass transfer data of Schafermeyer et al. (1975). In the latter experimental study, the average and instantaneous rates of mass transfer are presented for a droplet falling through a power law type of non-Newtonian continuous phase.

It was determined that when the Yamaguchi et al (1974) types of stream functions are used with the proposed mathematical model, the mass transfer results for the creeping flow regime and the results of the intermediate Reynolds number region converge. Comparison of the mass transfer results, obtaining by using different forms of stream functions, with the experimental results of Griffith (1960) suggests more successful application of the Nakano and Tien (1970) types of stream functions at higher Reynolds numbers ($10 < Re < 100$) and the Yamaguchi et al. (1974) types of stream functions at low Reynolds numbers ($Re < 10$).

The present model, when used with the appropriate stream functions, quantitatively describes the continuous phase mass transfer coefficient for a drop flowing through a power law type of fluid in the intermediate Reynolds number region as a function of the Reynolds number, the Peclet number, the viscosity ratio parameter, and the power law flow behavior index.

Theoretical and experimental studies have been published on the subjects of flow and mass transfer around a fluid droplet. In an early theoretical study of Hadamard (1911), a solution was obtained to the problem of fluid flow around an internally circulating droplet in the creeping flow regime. Taylor and Acrivos (1964) theoretically investigated the fluid dynamics of both a slightly deformed drop and a spherical drop by taking the inertial terms into account through the use of a singular perturbation method for low Reynolds number. Hirose and Moo-Young (1969) examined the fluid mechanics and mass transfer process for gas bubbles moving through a power law type of non-Newtonian fluid in the creeping flow regime. Nakano and Tien (1968) and Mohan (1974) obtained approximate solutions for the creeping flow of a power law fluid over a Newtonian fluid sphere for a wide range of viscosity ratio parameters and flow behavior indexes.

There have also been investigations in which the range of applicability of the velocity fields has been extended to Reynolds numbers above the creeping flow region. Hamielec and Johnson (1962) and Hamielec et al (1963) obtained, by means of Galerkin's method, the approximate stream functions for the movement of a droplet in a viscous Newtonian fluid in the intermediate Reynolds number range ($5 \lesssim Re' \lesssim 40$). Hamielec and co-workers (Hamielec et al., 1967; LeClair et al., 1972; Abdel-Alim and Hamielec, 1975) have numerically solved the Navier-Stokes equations for the stream functions inside and outside dispersed phases at Reynolds numbers significantly above the creeping flow regime and have applied their results to the calculation of drag coefficients. All of the above studies in the intermediate Reynolds number region have been for Newtonian fluids. Nakano and Tien (1970) extended the earlier analysis of Hamielec and co-workers for the flow of a drop in a Newtonian continuous phase in the intermediate Reynolds number region to include a power law type of continuous phase, also using Galerkin's method. More recently, Yamaguchi et al. (1974) obtained approximate solutions for the laminar flow of a fluid over a droplet. They used the Galerkin's method; however, they chose a different form of the stream functions which enabled them to predict a smooth transition for the drag coefficient and mass transfer coefficient from the creeping flow regime to the intermediate Reynolds number region. The analysis of Yamaguchi et al. involved only Newtonian fluids; however, their analysis can be extended to include a power law type of continuous phase.

Baird and Hamielec (1962) investigated the forced convective mass transfer around spheres at intermediate Reynolds numbers for the case of a thin concentration boundary layer ($Pe >> 1$) in Newtonian fluids. Wellek and Huang (1970) used the Nakano and Tien (1968) stream functions to numerically solve the diffusion equation to predict the continuous phase mass transfer coefficient for the creeping flow of a Newtonian fluid droplet in a power law type of fluid. Recently, Gürkan and Wellek (1976) reconsidered the above problem and used the more realistic Mohan (1974) stream functions for power law fluids. The problem of unsteady state mass transfer to fluid spheres in non-Newtonian continuous phases at high Reynolds number has been solved with some approximations by Shirotsuka and Kawase (1973). Mass transfer from gas bubbles to slightly non-Newtonian liquids that is accompanied by a chemical reaction has been analyzed by Dang et al. (1972) for the low Reynolds number region. Skelland and Ramanan (1975) applied the boundary layer approach to systems in which the continuous phase is non-Newtonian. They considered a continuous phase which obeys the power law model and extended the boundary-layer analysis approach of Griffith (1960). Skelland and Ramanan's results are applicable either in the intermediate or high Reynolds number regions, provided the Peclet number is large.

There have been a number of experimental mass transfer studies in which both of the phases are Newtonian, for example, Griffith (1960). Some of the more important investigations are summarized by Kintner (1963), Johns et al. (1965), Pritchard and Biswas (1967), Tavlarides et al. (1970), Heertjes and DeNie (1971), and Skelland (1971). Experimental studies on mass transfer from gas bubbles in non-Newtonian solutions have been reported by Barnett et al. (1966), Calderbank (1967), and Calderbank et al. (1970). Experimental studies have also been reported by Skelland and Ramanan (1975) and Schafermeyer et al. (1975) on liquid-liquid dispersed phase systems in which the continuous phase is non-Newtonian.

Up to the present time, there is no existing relationship for the continuous phase mass transfer coefficient which may be used in the intermediate Reynolds number region for the flow of a power law type of fluid over a Newtonian fluid sphere for the range of Peclet numbers from 1 to 10^6. This problem is solved in the present work by making use of the Nakano and Tien (1970) stream functions. Since the form of the stream functions which are used with the related fluid flow problem is expected to effect the mass transfer results to a significant degree, the results obtained by using the Nakano and Tien types of stream functions are compared to the results obtained by using the Yamaguchi et al. (1974) types of stream functions for the special case of Newtonian fluids. The results of this comparison will aid in future studies of flow and mass transfer in dispersed phases when one of the phases is non-Newtonian in character.

Before concentrating on the analysis, discussion, and conclusions presented in this work, it is perhaps of importance to indicate in a general manner one approach to the utilization of diffusional rate relations for dispersed phase systems. A procedure has been developed and successfully applied in the design of perforated plate extraction columns for Newtonian liquid-liquid systems by Skelland and Conger (1973). The method focuses on the calculation and use of a pseudoequilibrium curve. If sufficient empirical correlations or theoretical relations are available for the prediction of the contact areas and mass transfer coefficients during the formation, rise, and coalescence periods, then this procedure can be easily applied. If the continuous phase in the extraction system is non-Newtonian, the results of the present investigation and others (Gürkan and Wellek, 1976) can be incorporated into the Skelland and Conger procedure. Once the droplet velocity, droplet size, densities, and rheological properties of the liquid phases are known, the mass transfer coefficient can be calculated in the manner to be described in this article. The terminal velocity of the fluid spheres in non-Newtonian media can be predicted either by existing empirical correlations (Mhatre and Kintner, 1959; Fararoui and Kintner, 1961; Marrucci et al., 1970; Mohan et al., 1972) or theoretical relations (Nakano and Tien, 1968, 1970; Shirotsuka and Kawase, 1973; Mohan, 1974). The power law flow behavior index n and the consistency index K of the continuous phase can be easily measured in the laboratory along with the densities of both phases. Predictive theories can be used to estimate the molecular diffusion coefficient in non-Newtonian media (Clough et al., 1962; Hoshino, 1971; Navari et al., 1971), or experimental measurements can be used to obtain the diffusivity as has been done by Astarita (1965), Quinn and Blair (1967), Osmers (1969), Zandi and Turner (1970), and Wasan et al. (1972). The drop size in non-Newtonian liquids can be estimated by using the correlation of Skelland and Raval (1972).

Therefore, when the physical characteristics of the particular extraction system are determined, one can determine the Peclet number Pe, the Reynolds number Re, the viscosity ratio parameter X, and the flow behavior index n. The continuous phase mass transfer coefficients can then be determined, as outlined in this paper, and this information utilized in the Skelland and Conger (1973) design procedure.

THEORY

Forced convective mass transfer to a spherical bubble or droplet is described by the following dimensionless partial differential equation:

$$V_y \frac{\partial C}{\partial y} + \frac{V_\theta}{y}\frac{\partial C}{\partial \theta} = \frac{2}{Pe}\left[\frac{\partial^2 C}{\partial y^2} + \frac{2}{y}\frac{\partial C}{\partial y}\right] \qquad (1)$$

Spherical coordinates, which have their origin at the center of the sphere, are used. The coordinate system is chosen so that $\theta = 0$ corresponds to the frontal stagnation point. The following boundary conditions are used with the above equation:

$$C(1, \theta) = 0 \qquad (2a)$$

$$C(\infty, \theta) = 1 \qquad (2b)$$

$$\frac{\partial C(y, 0)}{\partial \theta} = 0 \qquad (2c)$$

The details of the mathematical model and related assumptions are given by Gürkan and Wellek (1976) who used a similar model to describe mass transfer to droplets in the creeping flow regime.

The relations, which approximate the velocity components V_θ and V_y, are derived in the present work from the Nakano and Tien (1970) intermediate Reynolds number stream functions. The use of these velocity components in Equation (1) is based on the assumption of uncoupled mass and momentum transfer mechanisms. The velocity components are given by the following equations:

$$V_y = (-1 + 2A_1y^{-3} + 2A_2y^{-4} + 2A_3y^{-5}) \cos\theta + (B_1y^{-3} + B_2y^{-4} + B_3y^{-5})(2\cos^2\theta - \sin^2\theta) \qquad (3a)$$

$$V_\theta = (1 + A_1y^{-3} + 2A_2y^{-4} + 3A_3y^{-5}) \sin\theta + (B_1y^{-3} + 2B_2y^{-4} + 3B_3y^{-5}) \cos\theta \cdot \sin\theta \qquad (3b)$$

The coefficients A_i and B_i ($i = 1, 2, 3$) are given as functions of the flow behavior index n, the Reynolds number Re, and the viscosity ratio parameter X in Table 1.*

The velocity components based on the Yamaguchi et al. (1974) low Reynolds number stream functions for a Newtonian continuous phase are also derived in the present work. They are given by

$$V_y = (-1 - 2E_1y^{-1} - 2E_2y^{-2} - 2E_3y^{-3} - 2E_4y^{-4}) \cos\theta + (-F_1y^{-1} - F_2y^{-2} - F_3y^{-3} - F_4y^{-4})(2\cos^2\theta - \sin^2\theta) \qquad (4a)$$

$$V_\theta = (1 + E_1y^{-1} - E_3y^{-3} - 2E_4y^{-4}) \sin\theta + (F_1y^{-1} - F_3y^{-3} - 2F_4y^{-4}) \cos\theta \cdot \sin\theta \qquad (4b)$$

The coefficients E_i and F_i ($i = 1, 2, 3, 4$) are given as functions of the Reynolds number Re' and the viscosity ratio R by Yamaguchi et al., (1974).

Equations (1) and (2) are numerically solved together with Equations (3) or (4) to obtain the solute concentration profiles in the continuous phase. The details of the solution of the mathematical mass transfer model by the Crank and Nicolson (1947) implicit numerical finite-difference technique are given by Wellek and Huang (1970) and by Gürkan (1976). The rate of mass transfer is represented and calculated by

$$Sh = \frac{k_c 2a}{D} = \int_0^\pi \left(\frac{\partial C}{\partial y}\right)_{y=1} \sin\theta \, d\theta \qquad (5)$$

In this relation, k_c is the average of the local mass transfer coefficients over the entire outer surface of the sphere.

The Nakano and Tien (1970) intermediate Reynolds number stream functions are also used with the following short range diffusion equation of Baird and Hamielec (1962):

$$Sh = \left[\frac{2}{\pi} Pe \int_0^\pi (V_\theta)_{y=1} \sin^2\theta \, d\theta\right]^{1/2} \qquad (6)$$

The following expression for the continuous phase Sherwood number, which is applicable at high Peclet numbers, is then obtained:

$$Sh = \left(\frac{8}{3\pi}\right)^{1/2} (1 + A_1 + 2A_2 + 3A_3)^{1/2} (Pe)^{1/2} \qquad (7)$$

RESULTS

The mass transfer model represented by Equations (1) through (3) is numerically solved for various values

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of the Reynolds number, n, X, and the Peclet number. The ranges of these parameters are: 1 to 10^6 for the Peclet number, 0.01 to 2 for X, 0.6 to 1.0 for n, and 5 to 25 for the Re. The number of radial increments is set at a value of 16. Angular increments of 2 deg. are used. The value of Δy to be used for each value of the Peclet number is determined in the same manner as discussed by Gürkan and Wellek (1976). These values are given in Table 2.*

The rate of mass transfer, represented by the Sherwood number, is calculated from the concentration profiles, which are obtained from the numerical solution of the mass transfer model in which the Nakano and Tien (1970) stream functions are used. The complete results are presented in Table 3.* Some typical results are shown in Figures 1 and 2. In Figure 1, the Sherwood number is presented as a function of the Reynolds number in the intermediate Reynolds number region for values of X equal to 0.01, 0.1, and 1.0, respectively. Three values of the flow behavior index are used for a Peclet number of 1 000: 0.7, 0.8, and 1.0.

Figure 1 shows that the continuous phase mass transfer coefficient increases with an increase in the Reynolds number when all other variables are held constant. Figure 1 also shows that an increase in the Sh occurs when n decreases (that is, for increased pseudoplasticity). The amount of increase in Sh with a decrease in n and an increase in Re depends on X. The maximum enhancement is seen for X values of about 0.1. The Sherwood number is presented as a function of X in Figure 2 for a Peclet number of 1 000 and a Reynolds number of 25. Figure 2 clearly shows the effects of X and n on the rate of mass transfer. Five values of n are employed: 1.0, 0.9, 0.8, 0.7, and 0.6.

The increase in the value of the Sherwood number with a decrease in the value of n can also be seen in Figure 2. The dependence of the Sh on X changes its character as the value of n changes. For values of n between unity and 0.8, the Sherwood number decreases with an increase in X, and most of the change occurs for X values between 0.1 and 1. However, when the value of n is less than 0.8, the Sh slightly increases with an increase in X from 0.01 to 0.1, displays a maximum at X about 0.1, and sharply decreases with further increase in X. An analysis of the results of the numerical solution indicates that the first value of n for which the dependence of the Sh on X shows a maximum depends on the values of the other parameters, especially the Reynolds number. At lower Reynolds numbers, the dependence of the Sherwood number on X does not exhibit a maximum.

The results showing the effect of the Peclet number on the dependence of the Sh on X are shown in Figure 3. As expected, the Sherwood number increases with an increase in the Peclet number when all other variables are held constant. Gürkan and Wellek (1976) developed a solution for the variation of the Sherwood number with the Peclet number for creeping flow of a power law type of fluid over a Newtonian droplet. The Mohan (1974) stream functions, which are applicable for very low drop Reynolds numbers, were used in the above analysis. Because the dependence of the Sh on the Pe in the intermediate Reynolds number region qualitatively exhibits the same features as the dependence of the Sh on the Pe in the creeping flow regime, it will not be discussed further. Also shown in Figure 3 are the results obtained through the use of Equation (7), which is the relation that is obtained by inserting the Nakano and Tien (1970) velocity profiles into the short range diffusion equation of Baird and Hamielec (1962).

* See footnote on page 486.

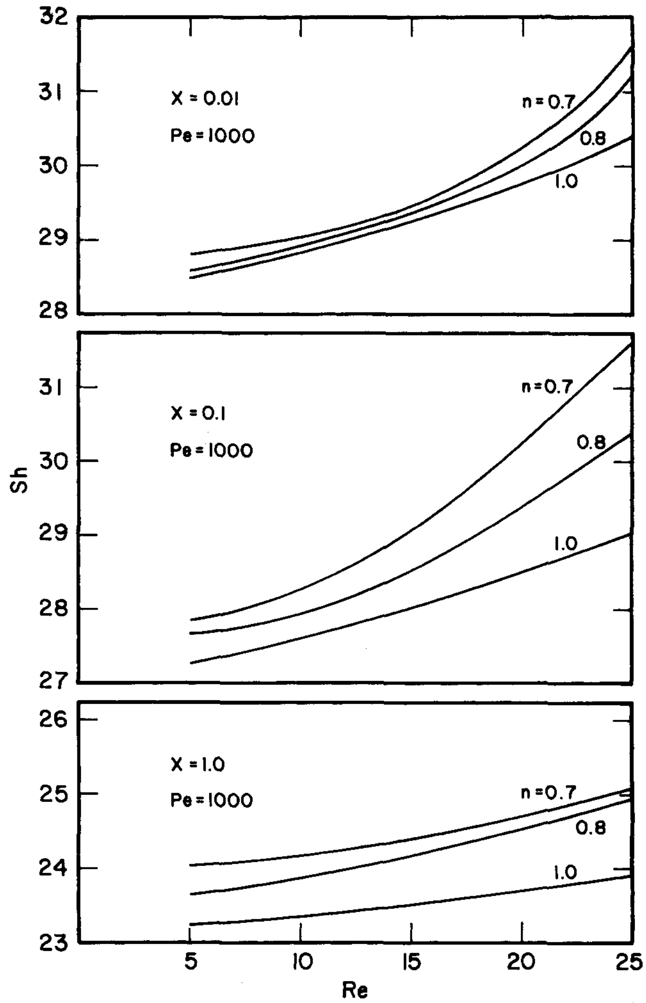

Fig. 1. The continuous phase Sherwood number as a function of the Reynolds number for parametric values of n, X, and Pe.

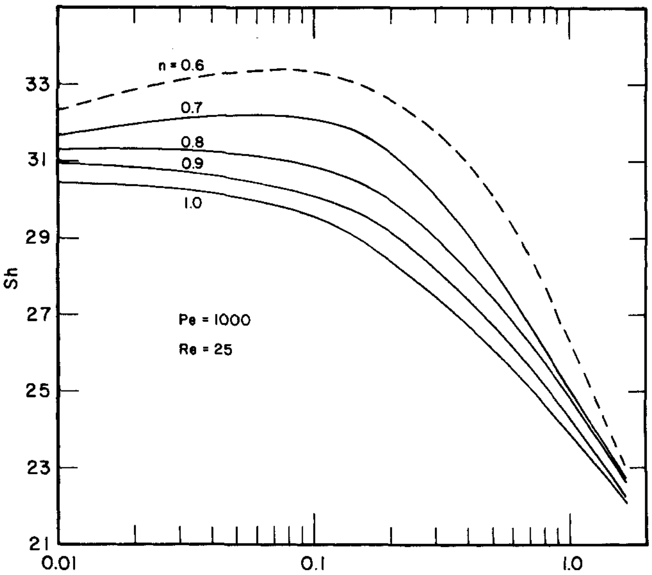

Fig. 2. The continuous phase Sherwood number as a function of the viscosity ratio parameter X for parametric values of n (Pe = 1 000; Re = 25).

DISCUSSION

Although the approximate nature of the Nakano and Tien (1970) stream functions causes some scatter in the results at high Peclet numbers and larger X values, it is

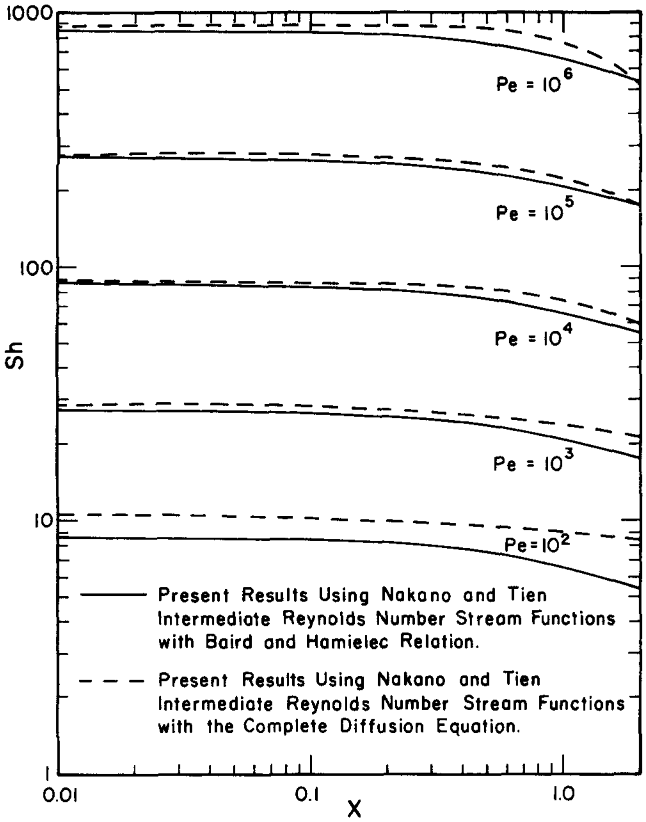

Fig. 3. A comparison of the Sherwood numbers calculated with the Baird and Hamielec (1962) relation and with the complete diffusion equation. ($Re = 10$; $n = 0.8$)

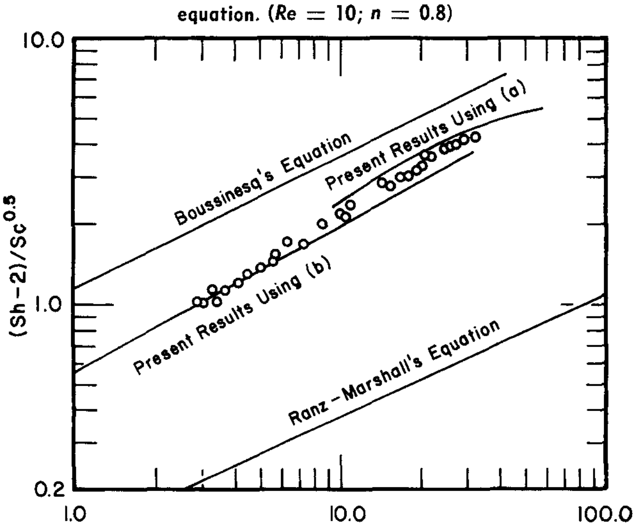

Fig. 4. Comparison of the experimental mass transfer rates of Griffith (1960) with the results calculated in the present work by using (a) the Nakano and Tien stream functions and (b) the Yamaguchi et al. stream functions for water drops in iso-butanol ($X = 0.38$; $Sc = 23\,600$; $n = 1.0$).

clear that in the intermediate Reynolds number region ($5 < Re' < 40$) the continuous phase Sherwood number increases with a decrease in n, the flow-behavior index. The same type of dependence of the Sh on n has been predicted in the creeping flow region ($Re' < 1$) by Gürkan and Wellek (1976) and Hirose and Moo-Young (1969). This has also been confirmed experimentally by Skelland and Ramanan (1975). However, in the high Reynolds number region, Shirotsuka and Kawase (1973) predicted an increase in the Sh with an increase in n. Their analysis also predicted an increase in the drag coefficient with an increase in n for the high Reynolds number region. In favorably comparing their predictions with the Nakano and Tien (1970) intermediate Reynolds number region drag coefficients, Shirotsuka and Kawase used only one set of data ($X = 2.0$) for $Re = 25$. The Nakano and Tien (1970) drag coefficients exhibit the opposite trend for all other values of X.

In Figure 3, a comparison of calculated drop Sherwood numbers is presented for two separate applications of the Nakano and Tien (1970) intermediate Reynolds number stream functions: inclusion in Equations (1) and (2), the complete diffusion relation; inclusion in Equation (6), the Baird and Hamielec (1962) short range diffusion relation. In an earlier study, Gürkan and Wellek (1976) found that when the Mohan (1974) creeping flow stream functions are used, the results obtained when the complete diffusion equation is solved are about the same as for the results obtained when the Baird and Hamielec relation is used for the ranges $Pe > 10^4$ and $X < 3$. This observation is also seen to apply in the intermediate Reynolds number region. This fact confirms the accuracy of the numerical solution.

The predictions of the present work are compared with experimental results in a recent paper by Schafermeyer et al., (1975). In this latter study, it is determined that the theoretical results of the present work, when applied at the same conditions as the experimental study, provide an upper bound to the mass transfer data.

As has been previously indicated, the mass transfer problem of the present work is also solved by using the low Reynolds number region stream functions of Yamaguchi et al. (1974) for the special case of a Newtonian continuous and dispersed phase. The results which are obtained by the use of the Yamaguchi et al. stream functions are compared with the results which are obtained by the use of the Nakano and Tien (1970) stream functions for the special case of n equal to unity. As may be seen in Figure 4, the comparison indicates that the former results were significantly lower than the latter. It has also been determined that the results obtained by using the Yamaguchi et al. stream functions converged smoothly with the creeping flow regime results of Gürkan and Wellek (1976). Yamaguchi et al. attribute the success of their stream functions to their special functional forms. The results of the calculations with the Yamaguchi et al. stream functions are shown in Table 4.*

The results obtained in this work are compared with the experimental data of Griffith (1960) in Figure 4. Also shown on the figure are the asymptotic results obtained by Boussinesq (1905) and Ranz and Marshall (1952) for a fully circulating and a stagnant drop, respectively. Figure 4 shows that the present results with the Nakano and Tien (1970) stream functions for n equal to unity compare more favorably with the experimental data for higher Reynolds numbers ($Re' > 10$), and the present results with the Yamaguchi et al. (1974) stream functions compare more favorably with the experimental data for lower Reynolds number ($Re' < 10$).

In conclusion, the above discussion for Newtonian fluids in both phases suggests that the Nakano and Tien (1970) types of stream functions can be used with considerable confidence for higher Reynolds number region ($10 < Re' < 100$) and Yamaguchi et al. (1974) types of stream functions for lower Reynolds number region ($Re' < 10$). Therefore, in future flow and mass transfer studies of both Newtonian and non-Newtonian dispersed phases, the appropriate stream functions should be chosen depending on the Reynolds number region within which one is working.

* See footnote on page 486.

NOTATION

a = spherical radius of dispersed phase, cm
A_i, B_i = coefficients in Equations (3a) and (3b)
c = concentration of the solute, moles/l
C = concentration of the solute, c/c_∞, dimensionless
D = diffusivity of solute, cm^2/s
E_i, F_i = coefficients in Equations (5a) and (5b)
k_c = continuous phase mass transfer coefficient, cm/s
K = consistency index of power law fluid; when $n = 1$, $K = \mu_c$
n = flow-behavior index of power law fluid
Pe = Peclet number, $2\,a\,V_\infty/D$, dimensionless
r = radial distance, cm
R = viscosity ratio, μ_d/μ_c, dimensionless
Re = Reynolds number $a^n V_\infty^{2-n}\,\rho_c/K$, dimensionless
Re' = Reynolds number, $(2a)^n V_\infty^{2-n}\,\rho_c/K$, dimensionless
Sc = Schmidt number $(K/\rho_c\,D)\,(V_\infty/2a)^{n-1}$, dimensionless
Sh = continuous phase Sherwood number, $k_c\,2a/D$, dimensionless
U_θ = tangential velocity, cm/s
U_r = radial velocity, cm/s
V_θ = tangential velocity, U_θ/V_∞, dimensionless
V_y = radial velocity, U_r/V_∞, dimensionless
V_∞ = relative velocity between sphere and continuous phase fluid, cm/s
X = viscosity ratio parameter, $(\mu_d/K)\,(a/V_\infty)^{n-1}$, dimensionless
y = radial displacement from center of sphere, r/a, dimensionless

Greek Letters

θ = angular displacement from front stagnation point, radians
μ = viscosity, poise
ρ = density, g/cm^3
τ_j^i = stress tensor
Δ_j^i = rate of deformation tensor
Δ_y = radial increment in finite difference equations
Δ_θ = angular increment in finite difference equations

Subscripts

c = continuous phase
d = dispersed phase
∞ = infinity

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Reaction of Sulfur Dioxide with Limestone and the Grain Model

Experimental measurements of the reaction of sulfur dioxide and oxygen with limestones have demonstrated substantial influence of the geologic origin of the stone, its porosity and particle size, gaseous concentration of sulfur dioxide, and temperature on the course of reaction and the conversion (that is, the degree of utilization of the limestone content of the particles as a sorbent for sulfur dioxide). A mathematical model including intraparticle transport and chemical reaction within the particles (grain theory) has been developed to simulate this sulfur dioxide sorption reaction.

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SCOPE

One of the more promising techniques of sulfur dioxide removal from flue gas is sorption using a fluidized bed of limestone. A major drawback in such processes, however, is the fact that the limestone reacts only partially. Although an extensive pore structure develops during calcination of the limestone, the sulfation reaction causes large decreases in porosity of the reacting particles (Borgwardt and Harvey, 1972; Hartman and Coughlin, 1974).

The purpose of the present study was to develop a better understanding of the intricacies of reaction involving limestone particles, sulfur dioxide, and oxygen, and to quantitatively describe the effects of particle size, gaseous sulfur dioxide concentration, and reaction time on the course of the chemical reaction.

Whereas previous investigations appear to have been largely confined to calcined limestone, the use of uncalcined limestone is a more practical expedient for fluidized-bed combustion. In the present work, uncalcined samples of reactive, high-grade limestone were exposed to flue gas in a differential, packed-bed reactor. The amount of sulfate formed in the samples was determined by titration with 0.005 N $Ba(ClO_4)_2$ and electron microprobe, and x-ray diffraction analyses were employed to study the spatial distribution of reaction within the particles. The effects of such variables as temperature, time of exposure to the gas, particle size, and gaseous concentration of sulfur dioxide were also determined.

The reaction model developed herein for this gas-solid reaction system takes into account the reduction in porosity caused by the sulfation reaction and the resulting effect on the rate of transport of sulfur dioxide through the pores. The development includes a quantitative description of the diffusion of sulfur dioxide through an accumulating shell of reaction product at the periphery of a limestone particle as well as the chemical reaction taking place at the active surface of the grains within the particle.

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