

01 Jan 1976

Mass Transfer In Dispersed And Continous Phases For Creeping Flow Of Fluid Spheres Through Power Law Fluids

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Recommended Citation

T. Gürkan and R. M. Wellek, "Mass Transfer In Dispersed And Continous Phases For Creeping Flow Of Fluid Spheres Through Power Law Fluids," *Industrial and Engineering Chemistry Fundamentals*, vol. 15, no. 1, pp. 45 - 52, American Chemical Society, Jan 1976.

The definitive version is available at <https://doi.org/10.1021/i160057a009>

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a_{ij}^0 = infinitely dilute a_{ij} with species j dilute
 a_{ij}^* = ideal a_{ij} defined by eq 12
 B = coefficient in eq 1
 D = self-diffusion coefficient
 D_{ij} = binary mutual diffusivity
 D_{ij}^0 = infinitely dilute D_{ij} with species j dilute
 F = self-friction coefficient
 F_{ij} = multi-component friction coefficient
 K = constant in eq 2
 R = gas constant
 S = pure liquid transport group, eq 5
 S_{ij} = mixture transport group, eq 8
 T = absolute temperature
 T_c = critical temperature
 T_r = reduced temperature, T/T_c
 V = molar volume
 V_c = critical volume
 V_0 = ultimate volume
 X = mole fraction

Greek Letters

α = activity coefficient
 β = thermodynamic factor = $1 + d \ln \alpha / d \ln X$
 γ = thermodynamic correction factor
 η = viscosity

θ = reduced mole fraction, eq 16
 φ = fluidity = $1/\eta$
 ψ = ultimate volume function, eq 13

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Received for review April 21, 1975

Accepted October 24, 1975

Mass Transfer in Dispersed and Continuous Phases for Creeping Flow of Fluid Spheres through Power Law Fluids

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The diffusion equation was numerically solved by an implicit finite-difference method for the purpose of calculating the continuous phase Sherwood number, Sh , for mass transfer from an internally circulating Newtonian droplet traveling through a non-Newtonian power-law-type continuous phase in the creeping flow regime. The Mohan stream functions were used in the calculations in order to approximate the velocity profile inside and outside the droplet. The calculated Sh is presented as a function of the Peclet number, Pe , power-law index, n , and a viscosity ratio parameter, X . Sh increases as n decreases in the pseudoplastic region. The dependence of Sh on n is important when Pe is greater than 10^2 , except when both $X > 1$ and $Pe > 10^4$. When used with the Mohan stream functions, the Baird and Hamielec short-range diffusion equation provides a close approximation for Sh when $X < 3$, provided that $Pe > 10^4$. The mass transfer model for the dispersed phase was also numerically solved in order to determine the effect of continuous phase pseudoplasticity, n . Although a slight increase in the total amount of mass transferred, \bar{A}_{mt} , with a decrease in n was determined, it is concluded that the power-law behavior in the continuous phase does not affect to any appreciable extent the internal mass transfer, either with or without chemical reaction in the fluid sphere. \bar{A}_{mt} increases with decreasing X , and this dependency is particularly important when mass transfer occurs with chemical reaction in the dispersed phase.

Introduction

Mass transfer from bubbles and drops for which both the continuous and dispersed phases are Newtonian in character has been extensively investigated because of its importance in chemical engineering processes. Recently, more attention has been given to the case in which the continuous phase is non-Newtonian. The types of non-Newtonian phases that are encountered in industrial operations are: polymer melts and solutions, such as rubber in toluene; lubrication oils; cellulose esters in organic solvents; elastomers; emulsions; paints; food products, such as apple sauce and ketchup; liquid sludge in activated sludge processes; and fermentation broth for penicillin production. Many of these systems require purifying operations in which inter-

phase mass transfer is important. In some cases, one of the phases is dispersed.

The problem of mass transfer from bubbles in non-Newtonian fluids was studied experimentally by Calderbank (1967), Barnett et al. (1966), and Calderbank et al. (1970). Wellek et al. (1975) and Ramanan (1973) experimentally studied mass transfer from liquid droplets in non-Newtonian fluids.

Theoretical studies of mass transfer to dispersed phases in non-Newtonian systems have also been made. Shirotzuka and Kawase (1973) used their own approximate stream functions to describe unsteady-state mass transfer to fluid spheres in non-Newtonian systems at high Reynolds numbers for both the dispersed and the continuous phases. Dang et al. (1972) considered the case of unsteady-state

continuous phase mass transfer when there is a chemical reaction between bubbles and slightly non-Newtonian liquids at low Reynolds numbers. Ramanan (1973) extended the boundary layer analyses of earlier workers (Griffith, 1960) to cases in which the continuous phase is non-Newtonian, and he obtained relationships describing the continuous phase mass transfer coefficient.

Hirose and Moo-Young (1969) developed a theoretical relation, which describes the continuous phase mass transfer mechanism for a fully circulating gas bubble that moves in the creeping flow regime through a non-Newtonian liquid continuous phase described by the power-law rheological model. Wellek and Huang (1970) numerically solved the diffusion equation for the purpose of calculating the continuous phase mass transfer coefficient for creeping flow of a Newtonian type gas bubble or liquid droplet in a power-law type, non-Newtonian fluid. In order to consider the wide range of fluid sphere to continuous phase viscosities, Wellek and Huang used the Nakano and Tien (1968) continuous phase stream functions.

In order to estimate fluid sphere drag coefficients more accurately, Mohan (1974) recently reconsidered the case in which a power-law fluid moves over a Newtonian sphere in the creeping flow regime. In the process, he established a new set of stream functions, which take into account the energy dissipation in both the continuous and dispersed phases. This is in contrast to Nakano and Tien (1968) who in their analyses had neglected the dispersed phase energy dissipation. Mohan's primary intention was to evaluate the true upper bound on sphere drag. A comparison of Mohan's results with those of Nakano and Tien show that the upper bound on drag calculated by Mohan is significantly lower, especially when values of the relative viscosity ratio parameter are approximately near unity. The comparison suggests that the use of the Nakano and Tien stream functions may produce errors in liquid-liquid extraction mass transfer studies.

A considerable quantity of experimental and theoretical studies involving mass transfer that occurs with and without chemical reaction inside single droplets and gas bubbles when both the dispersed and continuous phases are Newtonian has been published. Johns and Beckmann (1966) solved the diffusion equation for the purpose of calculating the amount of mass transfer inside fluid droplets. Wellek et al. (1970) and Watada et al. (1970) calculated the rate of mass transfer when a first-order chemical reaction occurs inside fluid spheres. Brunson and Wellek (1971) solved the problem of mass transfer that is accompanied by a second-order chemical reaction inside fluid spheres. No study which attempts to predict the effect of the non-Newtonian continuous phase on the dispersed phase mass transfer mechanism has been published.

The objectives of the present work are (1) to use the continuous phase stream functions of Mohan (1974) with the diffusion equation to obtain more accurate continuous phase mass transfer coefficients for a power-law type non-Newtonian continuous phase flowing over a Newtonian fluid sphere in the creeping flow regime, (2) to compare the mass transfer coefficients obtained by using the Mohan continuous phase stream functions to those obtained by using the Nakano and Tien (1968) stream functions, and (3) to determine the effect of a non-Newtonian continuous phase on the dispersed phase mass transfer mechanism when the dispersed phase is Newtonian.

Continuous Phase Mass Transfer Studies

Mass Transfer Model. The mathematical model for mass transfer to spherical droplets or bubbles moving in

the creeping flow regime is described by the following dimensionless partial differential equation and boundary conditions; spherical coordinates with the origin at the center of the sphere are used

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

The boundary conditions are

$$C_A = 0 \text{ at } y = 1 \quad (2a)$$

$$C_A = 1 \text{ at } y \rightarrow \infty \quad (2b)$$

$$\frac{\partial C_A}{\partial \theta} = 0 \text{ at } \theta = 0 \quad (2c)$$

The following assumptions are made. (1) The flow is axially symmetric and isothermal, and steady state has been reached. (2) The dispersed phase is a Newtonian fluid, whereas the continuous phase obeys a power-law model, i.e.

$$\tau_j^i = -\mu \Delta_j^i \quad (\text{for dispersed phase})$$

$$\tau_j^i = -K \left(\frac{1}{2} \Delta_k^m \Delta_k^m \right)^{(n-1)/2} \Delta_j^i \quad (\text{for continuous phase})$$

(3) The natural convection, the molecular diffusion in the angular direction, and the resistance to mass transfer inside the sphere are negligible. (4) The solute concentration is dilute, and the solute diffusivity and density of both phases are constant.

To complete the description of the forced-convection mass transfer model, the velocity components, V_θ and V_y , have to be specified. The following continuous phase velocity components are derived from the Mohan (1974) stream functions

$$V_y = (-1 + 2A_1 y^{\sigma-2} + 2A_2 y^{-3}) \cos \theta \quad (3a)$$

$$V_\theta = (1 - A_1 \sigma y^{\sigma-2} + A_2 y^{-3}) \sin \theta \quad (3b)$$

The coefficients, A_1 and A_2 , and the exponent, σ , are tabulated for different values of the flow behavior index, n , and a viscosity ratio parameter, X , which is defined in eq 4 as

$$X = \frac{\mu_d}{K} \left(\frac{a}{V_\infty} \right)^{n-1} \quad (4)$$

These coefficients were chosen by Mohan so that for the Newtonian case, $n = 1$, the Mohan stream functions would become identical with the Hadamard (1911) stream functions for continuous and dispersed phase Newtonian fluids.

The velocity components based on the Nakano and Tien (1968) stream functions are used in this work in order to reexamine the accuracy of the earlier work of Wellek and Huang (1970). The components given by Wellek and Huang are

$$V_y = (-1 + 2F_1 y^{\gamma-2} + 2F_2 y^{-3}) \cos \theta + (E_1 y^{-3} + E_2 y^{-4})(2 \cos^2 \theta - \sin^2 \theta) \quad (5a)$$

$$V_\theta = (1 - F_1 \gamma y^{\gamma-2} + F_2 y^{-3}) \sin \theta + (E_1 y^{-3} + 2E_2 y^{-4}) \cos \theta \sin \theta \quad (5b)$$

By using eq 3a and 3b, the boundary value problem containing eq 1 and 2 can be solved numerically for the solute concentration profiles in the continuous phase around the bubble or droplet. The continuous phase Sherwood number is then calculated as

$$\text{Sh} = \frac{k_c 2a}{D} = \int_0^\pi \left(\frac{\partial C_A}{\partial y} \right)_{y=1} \sin \theta d\theta \quad (6)$$

in which k_c is the average of the local mass transfer coefficients over the entire outer surface of the sphere. As al-

readily indicated, eq 5a and 5b are used with the boundary value problem described by eq 1 and 2. The details of the numerical solution of the boundary value problems in which the Crank-Nicolson (1947) implicit numerical method is used are given by Wellek and Huang (1970) and by Gürkan (1976).

When Mohan's (1974) continuous phase velocity profiles are inserted into the following short-range diffusion equation of Baird and Hamielec (1962)

$$\text{Sh} = \left[\frac{2}{\pi} \text{Pe} \int_0^\pi (V_\theta)_{y=1} \sin^2 \theta \, d\theta \right]^{1/2} \quad (7)$$

the following expression is obtained for the continuous phase Sherwood number

$$\text{Sh} = \sqrt{\frac{8}{3\pi}} (1 - A_1\sigma + A_2)^{1/2} \text{Pe}^{1/2} \quad (8)$$

The above equation is theoretically applicable only for fully circulating gas bubbles and liquid droplets; however, we have determined to what extent this restriction can be relaxed for moderately large values of the viscosity ratio parameter, X .

Results and Discussion

The boundary value problem represented by eq 1 and 2 for the forced-convective mass transfer system was solved numerically for various values of Peclet numbers ranging between 1 and 10^6 . The results are presented in terms of Sherwood numbers as a function of the Peclet number, the continuous phase flow behavior index, n , and the viscosity ratio parameter, X . The coefficients of the stream functions were presented by Mohan (1974) for discrete values of X and n , ranging from 0.001 to 1000 and 0.6 to 1.0, respectively. The values of the coefficients of the Mohan stream functions are presented in Table III. (See the paragraph at the end of the paper regarding supplementary material.)

Prior to obtaining the final numerical solution of the mathematical model, the number of radial and angular increments and the size of the radial and angular increments, Δy , and $\Delta\theta$ had to be determined. The number of radial increments was set at a value of 16. Angular increments of 2° were found suitable. A reduction of the angular increment by a factor of 2 did not result in a significant change in the calculated values of the Sherwood numbers.

As expected, we observed that as the Peclet number increases the concentration boundary layer becomes thinner. This necessitated the determination of a specific value of Δy for each value of the Peclet number. At each Peclet number, several values of Δy were used to calculate the Sherwood number at a certain set of values of X and n . An analysis of the results indicated that the calculated Sherwood number is a minimum at a certain value of the radial increment size, and the Sherwood number steadily increases for larger or smaller values of Δy . This value of Δy gives, in practically all cases, the most reasonable concentration profile. That is, only at this value of Δy , the concentration at the grid point farthest from the drop surface reaches a value of unity for θ equal to zero, which is the front stagnation point. Of course, at higher values of Δy the same condition is also met, but in these cases, the concentration profile is greatly suppressed. For smaller values of Δy , the round-off error associated with the finite-differences numerical method probably becomes more important as indicated by a sharp increase in the value of Sh . For larger values of Δy , the truncation error probably becomes more important as indicated by a gradual increase in values of Sh , the continuous phase Sherwood number. Therefore in our judgment, the minimal value of the Sh corresponds

to the minimum total computational error, and the corresponding value of Δy can be used for other values of X and n . The values of Δy that we used are presented in Table IV for each value of the droplet Peclet number. (See the paragraph at the end of the paper regarding supplementary material.)

The Mohan (1974) stream functions are applicable to either spherical liquid droplets or gas bubbles, which travel in the creeping flow regime through a non-Newtonian continuous phase described by a power-law type constitutive equation. The average continuous phase Sherwood number ($k_c 2a/D$) was calculated as a function of the sphere Peclet number, Pe , the power-law flow behavior index, n , and the viscosity ratio parameter, X . (See Table V in the supplementary material.) Mass transfer results, which use the Mohan (1974) stream functions, are presented as a function of Pe in Figure 1 for values of n equal to 1.0, 0.8, and 0.6, respectively. Four values of the viscosity ratio parameter, $X = (\mu_d/K)(a/V_\infty)^{n-1}$, are employed: 0.001, 1.0, 10, and 1000. The Sherwood number relations are also presented in Figure 2 in order to describe more clearly the effect of the viscosity ratio parameter, X , and the power-law index, n .

As can be seen in Figure 1, the Sherwood number increases as the Peclet number increases for both non-Newtonian and Newtonian continuous phases. The droplet mass transfer rates are less dependent on the Peclet number when it is less than about 10^2 .

The effect of the power-law index, n , on mass transfer in the continuous phase becomes important when the Peclet number is greater than about 100. As n decreases from unity, the Sherwood number increases; i.e., increased pseudoplasticity results in an enhancement of mass transfer when all other variables are held constant. Although the dependence of the Sherwood number on n is important for Peclet numbers larger than 10^2 , this dependence becomes smaller as the viscosity ratio parameter becomes larger than unity. The region in which the variation of Sh with n is important for all values of X occurs for Peclet numbers around 100. For $X \approx 0.001$, the dependence of Sh on n seems to be nonlinear. For example, about two-thirds of the increase in the value of the Sherwood number occurs when n decreases from 1.0 to 0.80.

The dependency of the continuous phase Sherwood number on the viscosity ratio parameter, X , is noticeable when $\text{Pe} \geq 5$, and the effect is very pronounced when the Peclet number is greater than about 100. As can be seen in Figure 2, the Sherwood number is most sensitive to changes of X for values of the viscosity ratio parameter between 0.1 and 10.0. The effect of the viscosity ratio parameter asymptotically diminishes as X approaches either 0.001 or 1000. These asymptotes reflect the behavior of the "fully" circulating and the slightly circulating droplet systems, respectively.

Although it is expected that the Mohan (1974) stream functions are superior to those of Nakano and Tien (1968), the earlier calculations of Wellek and Huang (1970) and Hirose and Moo-Young (1969) were reexamined. These results are then compared with the results presented above.

Hirose and Moo-Young (1969), in a study of drag and mass transfer characteristics of a spherical gas bubble moving in creeping flow through a power-law non-Newtonian fluid, derived the following continuous phase velocity components

$$V_y = \left\{ \left[1 - \left(\frac{1}{y} \right) \right] - \frac{6n(n-1)}{(2n+1)} \left[\frac{1}{y} \ln y + \frac{1}{6} \left(\frac{1}{y} \right)^3 - \frac{1}{6} \left(\frac{1}{y} \right) \right] \right\} \cos \theta \quad (9a)$$

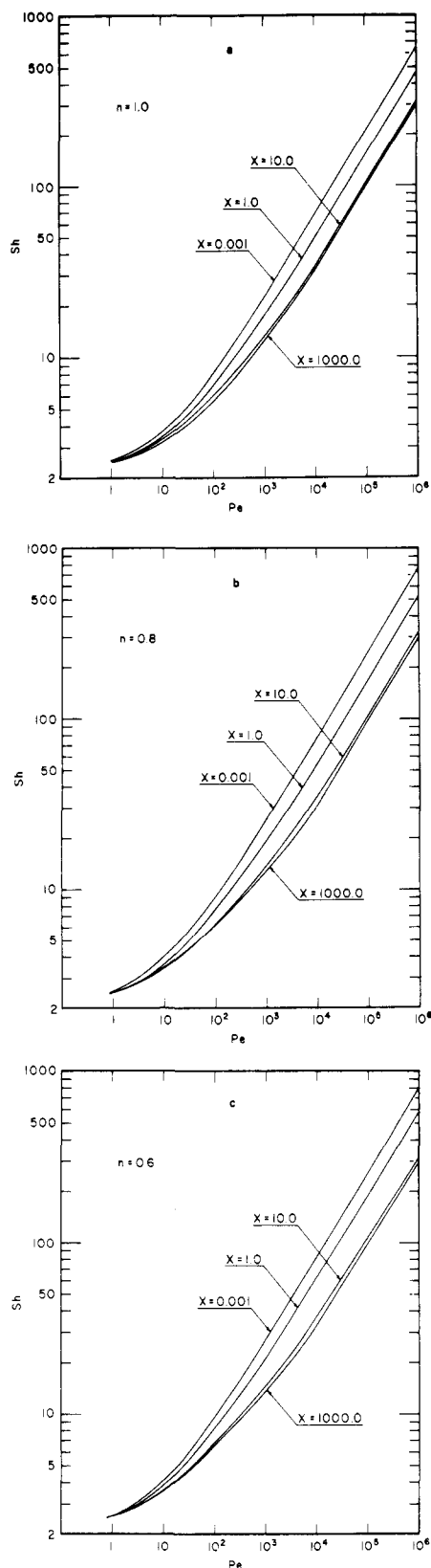


Figure 1. Sherwood number as a function of the Peclet number and the viscosity ratio parameter, based on Mohan (1974) stream functions. (a) $n = 1.0$; (b) $n = 0.8$; (c) $n = 0.6$.

$$V_{\theta} = - \left\{ \left[1 - \frac{1}{2} \left(\frac{1}{y} \right) \right] - \frac{3n(n-1)}{(2n+1)} \left[\frac{1}{y} \ln y - \frac{1}{5} \left(\frac{1}{y} \right)^3 + \frac{5}{6} \left(\frac{1}{y} \right) \right] \right\} \sin \theta \quad (9b)$$

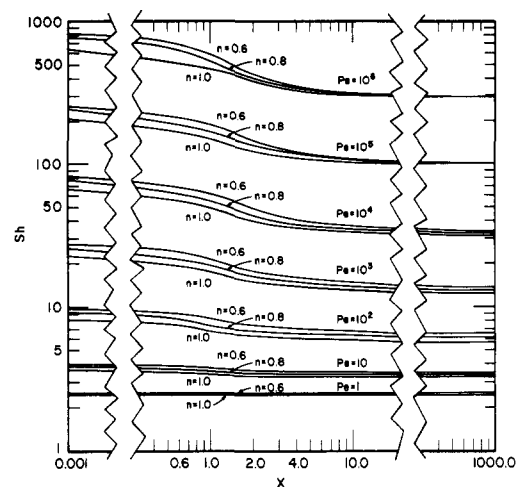


Figure 2. Effect of the viscosity ratio parameter X on Sherwood number for representative values of power-law index, n , and Pe , based on Mohan (1974) stream functions.

In their derivation of the above relations, it was assumed that the gas phase can be regarded as inviscid relative to the external liquid phase (i.e., $X \approx 0$) and that the deviation from Newtonian behavior is small. Upon application of Baird and Hamielec's (1962) relation for short-range diffusion from fully circulating spherical bubbles, Hirose and Moo-Young obtained the following relation between the Sherwood number, Peclet number, and power-law index, n

$$Sh = 0.65 \left[1 - \frac{4n(n-1)}{(2n+1)} \right]^{1/2} Pe^{1/2} \quad (10)$$

The viscosity ratio parameter, X , is not considered in the above derivation. Hence, situations in which the dispersed phase viscosity is not negligible compared with the continuous phase non-Newtonian fluid will probably not be properly described by eq 10, as for example, in the case of liquid-liquid extraction.

Wellek and Huang (1970) numerically solved the boundary value problem (eq 1 and 2 of this work) by using the Nakano and Tien (1968) stream functions. Their results agree with those of Hirose and Moo-Young (1969) up to a Peclet number of about $5 \times (10)^3$ for very small values of the viscosity ratio parameter ($X \approx 0.001$). However, for higher values of the Peclet numbers, their results are consistently lower than the results of Hirose and Moo-Young. They were unable to determine the reason for the deviation at high Peclet numbers. Moo-Young and Hirose (1972) later showed that the Sherwood number expression, which they had developed by use of the thin-boundary-layer approximation, for bubbles in power-law non-Newtonian fluids (i.e., eq 10) gives the minimum value for the Sherwood number.

We have determined that the relatively large values of the radial increment size used by Wellek and Huang (1970) probably account for the previously observed deviation, at higher Peclet numbers, between their work and that of Hirose and Moo-Young (1969). At higher Peclet numbers, the concentration gradient near the drop's surface is very steep. Wellek and Huang (1970) used a linear approximation to calculate the concentration gradient at the interface. This linear approximation, coupled with a large radial increment size, underestimated the continuous phase concentration gradient at the surface and, thus, underestimated the Sherwood number. In the present work, smaller values for the radial increment sizes have been used, and the concentration profile has been approximated by a second order curve-fit near the interface. The derivative of

Table I. Comparison of the Numerical Results Obtained by Using Mohan Stream Functions with the Results Using Nakano and Tien Stream Functions

X	n	(Sh) _{NT} /(Sh) _M			
		Pe = 1	Pe = 10 ²	Pe = 10 ⁴	Pe = 10 ⁶
0.001	1.0	1.0	1.0	1.0	1.0
1.0	1.0	1.0	1.0	1.0	1.0
10.0	1.0	1.0	1.0	1.0	1.0
0.001	0.9	1.00158	1.02663	1.05629	1.3076
1.0	0.9	1.00381	1.06826	1.09418	1.09816
10.0	0.9	1.00081	1.01455	1.00960	1.00544
0.001	0.8	1.00033	1.00482	1.00594	1.00606
1.0	0.8	1.00581	1.09911	1.16010	1.47089
10.0	0.8	1.00249	1.04169	1.03727	1.07750
0.001	0.7	1.00164	1.02345	1.02875	1.02931
1.0	0.7	1.00491	1.07994	1.10778	1.11184
10.0	0.7	1.00230	1.03739	1.03410	1.01991
0.001	0.6	1.00257	1.03620	1.02633	0.85336
1.0	0.6	1.00276	1.04220	1.05345	1.05469
10.0	0.6	1.00303	1.04841	1.04454	1.02372

this second-order curve was determined at the interface, and this derivative has in turn been used to calculate the Sherwood number. These corrected results are denoted as (Sh)_{NT} in the following comparison of results for continuous phase Sherwood numbers for which different stream functions are used.

The present results in which the stream functions of Mohan (1974) have been used are compared with those of Hirose and Moo-Young (1969) for circulating gas bubbles (see Figure 3). As can be seen in the figure, the Hirose and Moo-Young results are consistently lower, and the two curves approach each other at higher Peclet numbers as expected.

A comparison of the Sherwood numbers obtained by using the Mohan (1974) and the Nakano and Tien (1968) velocity profiles is shown in Table I for different values of X, n, and Pe. Both of these velocity profiles were originally derived so that they would converge with the Hadamard (1911) velocity profiles for n equal to unity. Thus, a value of unity for Sh_{NT}/Sh_M is expected for n equal to unity; this was obtained. As already pointed out, Mohan's analysis includes the internal circulation effects; thus, one should expect lower Sherwood numbers when his velocity profiles are used. It can be seen in Table I that this is always true, except at one point (Pe = 10⁶; X = 0.001; n = 0.6) where we have reason to believe that the Nakano and Tien (1968) stream functions are in error. It is seen in Table I that the differences in continuous phase Sherwood numbers can be quite important at higher Peclet numbers. The results we obtained by using the Mohan velocity profiles are smoother than those obtained with the Nakano and Tien stream functions. At every value of the Peclet number, when the value of n decreased, the value of the Sherwood number increased. This is an expected trend and is confirmed by several workers (Hirose and Moo-Young, 1969; Ramanan, 1973).

Detailed analysis of the results indicates that as X increases the differences with respect to n decrease, especially at high Peclet numbers. This condition can best be seen in Figure 2. One can observe that as the Peclet number increases the concentration boundary layer becomes thinner. The concentration gradient becomes restricted to a very small distance from the drop's surface, and the degree of mobility of the interface becomes a major factor. The interface mobility effect becomes so important that the variation with n is not detectable at higher Peclet numbers, particularly when the viscosity ratio parameter, X, is larger than 10.

It should be recalled that in this analysis molecular dif-

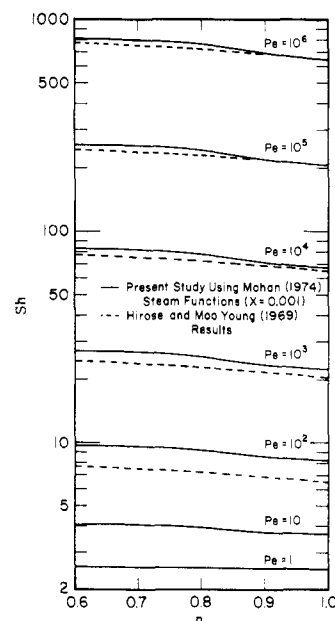


Figure 3. Effect of non-Newtonian behavior on mass transfer from circulating gas bubbles. Comparison of present work with Hirose and Moo-Young (1969) results.

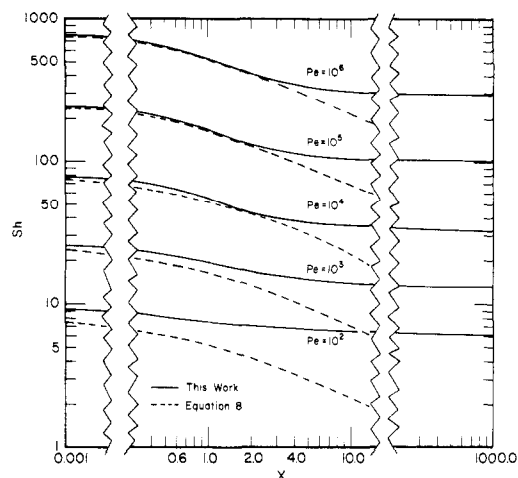


Figure 4. Effect of the viscosity ratio parameter X on Sherwood number. Comparison with eq 8, n = 0.8.

fusion in the angular direction has been neglected. The importance of diffusion in the angular direction increases as Pe approaches a value of unity. For example, if one compares the value of Sh predicted in this work (at Pe = 1 and n = 1) with those values obtained by Yuge (1956), Woo and Hamielec (1971), Dennis et al. (1973), and Oelrich et al. (1973), who have taken into account the effect of the angular diffusion component, the analysis in this work is found to overestimate Sh by about 9%. However, this effect rapidly loses its importance for Peclet numbers equal to or greater than 10.

Equation 8, which we developed primarily for fully circulating gas bubbles and liquid droplets, was examined to determine its usefulness at higher values of the viscosity ratio parameter, X. A comparison of the results obtained by eq 8 with the results of the boundary value problem is shown in Figure 4 for n = 0.8 and Peclet numbers larger than 10². This figure is representative of results obtained for the other values of n that have been used in this work. Figure 4 indicates that although eq 8 has been derived for fully circulating gas bubbles and liquid droplets (X ≤ 0.01) it can

be safely used for X values up to about 3, provided Peclet numbers are larger than about 10^4 . The mass transfer rate in the continuous phase can be estimated with eq 8 for different values of X and n , if the coefficients A_1 , A_2 , and σ are correlated as a function of X and n .

Wellek and Huang (1970) compared their results with other works when the continuous phase is Newtonian. They concluded that their results agree well with the various solutions of previous workers up to Peclet numbers of about 10^3 and that they were lower than the results of other studies for Peclet numbers greater than 10^3 . However, the results obtained in the present work compare favorably with the results of previous workers, all of whom used boundary layer analyses, and the values of the Sherwood numbers obtained are higher than those of other workers even at Peclet numbers of 10^4 . We conclude that the discrepancy between the Wellek and Huang results and the boundary layer analyses presented in the literature for the Newtonian continuous phase when the Peclet numbers are around 10^4 occurred because of the reasons which were found to be applicable in the discussion of the discrepancy between the Hirose and Moo-Young (1969) results and the Wellek and Huang results when the continuous phase is non-Newtonian. Therefore, the results of the present work, which are an improvement over the Wellek and Huang results especially for higher Peclet numbers, can be used to calculate the continuous phase mass transfer coefficient when the flow behavior is either Newtonian or non-Newtonian.

Dispersed Phase Mass Transfer Studies

Mass Transfer Model. The mathematical model for mass transfer with second-order chemical reaction inside a circulating fluid sphere which travels in the creeping flow regime can be described by the following dimensionless differential equations and initial and boundary conditions

$$\frac{\partial C_A}{\partial T} = \frac{\partial^2 C_A}{\partial y^2} + \frac{2}{y} \frac{\partial C_A}{\partial y} + \frac{1}{y^2} \frac{\partial^2 C_A}{\partial \theta^2} + \frac{\cot \theta}{y^2} \frac{\partial C_A}{\partial \theta} - \frac{1}{2} \text{Pe} \left(V_y \frac{\partial C_A}{\partial y} + \frac{V_\theta}{y} \frac{\partial C_A}{\partial \theta} \right) - k_R C_A C_B \quad (11a)$$

$$\frac{\partial C_B}{\partial T} = R_D \left[\frac{\partial^2 C_B}{\partial y^2} + \frac{2}{y} \frac{\partial C_B}{\partial y} + \frac{1}{y^2} \frac{\partial^2 C_B}{\partial \theta^2} + \frac{\cot \theta}{y^2} \frac{\partial C_B}{\partial \theta} \right] - \frac{1}{2} \text{Pe} \left[V_y \frac{\partial C_B}{\partial y} + \frac{V_\theta}{y} \frac{\partial C_B}{\partial \theta} \right] - R_C k_R C_A C_B \quad (12a)$$

The initial conditions are

$$C_A(0, y, \theta) = 0 \quad (11b)$$

$$C_B(0, y, \theta) = 1 \quad (12b)$$

and the boundary conditions are

$$\frac{\partial C_A}{\partial y}(T, 0, \theta) = 0 \quad (11c)$$

$$\frac{\partial C_B}{\partial y}(T, 0, \theta) = 0 \quad (12c)$$

$$C_A(T, 1, \theta) = 1 \quad (11d)$$

$$\frac{\partial C_B}{\partial y}(T, 1, \theta) = 0 \quad (12d)$$

$$\frac{\partial C_A}{\partial \theta}(T, y, 0) = 0 \quad (11e)$$

$$\frac{\partial C_B}{\partial \theta}(T, y, 0) = 0 \quad (12e)$$

$$\frac{\partial C_A}{\partial \theta}(T, y, \pi) = 0 \quad (11f)$$

$$\frac{\partial C_B}{\partial \theta}(T, y, \pi) = 0 \quad (12f)$$

The following assumptions are made. (1) All solutions are dilute so that the flow and diffusion equations are uncoupled. (2) The dispersed phase is a viscous sphere in linear or creeping motion. (3) The continuous phase resistance to solute diffusion is negligible. (4) Heat of reaction is low enough so that heat effects do not cause variations in any of the physical properties of the solutions. (5) The dispersed phase is a Newtonian fluid, whereas the continuous phase obeys a power-law model.

To complete the description of the model, the velocity components, V_θ and V_y , were derived for the present work from the Mohan (1974) stream functions

$$V_y = 2K_1(y^2 - 1) \cos \theta \quad (13a)$$

$$V_\theta = 2K_1(1 - 2y^2) \sin \theta \quad (13b)$$

Equations 11 and 12 were solved by an explicit finite-difference numerical method. The numerical technique used is similar to that used by Brunson and Wellek (1971), and details concerning the method are contained in that reference and in Gürkan (1976).

Results and Discussion

Equations 11a and 12a were numerically solved with the initial and boundary conditions given in eq 11b-f and 12b-f. The total amount of mass transferred, \bar{A}_{mt} , was calculated as a function of the time of contact, T , n , X , Pe , R_D , R_C , and k_R . For most computer solutions of the finite difference equations, the increments used were $\Delta T = 2 \times 10^{-4}$, $\Delta y = 1/40$, and $\Delta \theta = \pi/31$. To perform calculations for Peclet numbers as high as 1000, the angular and radial increments were kept as originally stated, and the dimensionless time increment was reduced to 5×10^{-5} .

When the reaction number is 0 and the flow behavior index is 1, the solution reduces to mass transfer without chemical reaction for a Newtonian continuous phase as presented by Johns and Beckmann (1966). This reference solution was used as a check for the computer program of this section.

The ranges of the variables considered in the study are given in Table II. The complete results of the computations are given by Gürkan (1976). Because the dependence of \bar{A}_{mt} on variables Pe , R_D , R_C , and k_R is discussed in detail in an earlier study (Brunson and Wellek, 1971), the discussion will not be repeated here.

The main objective of this portion of the present investigation was to determine the effects of the continuous phase flow behavior index, n , and the viscosity ratio parameter, X , on the dispersed phase mass transfer mechanism. Typical results showing these effects are given in Figure 5 and Figure 6. Figure 5 indicates that the variation of \bar{A}_{mt} with n is a function of the dimensionless second-order reaction rate constant, k_R . The maximum variation of \bar{A}_{mt} with n occurs when k_R has a value of about 40. This is the region where both the diffusion effects and the chemical reaction effects are important. Figure 5 shows that a decrease in the value of the flow behavior index, n (increased pseudoplasticity in the continuous phase), results in an increase in the total amount of mass transferred, \bar{A}_{mt} , when all other variables are held constant. Figure 6 indicates that the variation of \bar{A}_{mt} with X is also a function of k_R . In this case, however, the variation of \bar{A}_{mt} with X is important for values of $k_R > 0$. Figure 6 clearly shows that as the viscosity ratio parameter decreases the total amount of mass transferred, \bar{A}_{mt} , increases when all other variables are held constant.

Table II. Values of Variables Used for The Dispersed Phase Mass Transfer Studies

R_D	1.0			
R_C	0.2			
n	0.6	0.9	1.0	
X	0.001	1.0	10	
Pe	1	400	800	1000
k_R	0	40	160	640

The results of the calculations indicate that the maximum variation of \bar{A}_{mt} with n occurs for Peclet numbers of about 400 and also for X values around unity when k_R values are about 40. The variation of \bar{A}_{mt} with n was very much suppressed for X values of 10 except when the second-order reaction constant k_R values were around 640. It was not possible to go to values of the Peclet number higher than 1000 for $X \leq 1$ because of instability problems in the computer calculations.

The results of the calculations revealed that the total amount of mass transferred, \bar{A}_{mt} , inside the droplet varies very little with the continuous phase flow behavior index, n , for all combinations of the variables studied. The effect of the viscosity ratio parameter, X , on the total amount of mass transferred is important, however, except for the case of mass transfer without chemical reaction, $k_R = 0$. It should be stressed that these results are based on the assumption of the validity of the Mohan (1974) stream functions.

It should be noted that when the value of the second-order reaction rate constant, k_R , is set equal to zero the problem reduces to mass transfer without chemical reaction inside the droplet. The results of the solution for $k_R = 0$ can be used to calculate the total amount of mass transferred in the case of mass transfer with a first-order chemical reaction when the superposition method (Danckwerts, 1951; Stewart, 1968; Wellek et al., 1970) is used. Therefore, the conclusion that the effect of the continuous phase non-Newtonian behavior on the dispersed phase mass transfer mechanism is insignificant is general, and it covers mass transfer and mass transfer with first- and second-order reaction inside the droplet.

Conclusions

The analysis of mass transfer rates in the pseudoplastic continuous phase leads to the following conclusions.

(1) The Sherwood number, Sh , calculated using the Mohan (1974) stream functions, increases as the power-law flow behavior index, n , decreases in the pseudoplastic region. Although the dependence of Sh on n is important for sphere Peclet numbers, Pe , larger than 10^2 , this dependence becomes smaller as the viscosity ratio parameter, X , becomes larger than unity for $Pe > 10^4$. The variation of Sh with n is important for all values of X when Pe has values around 100.

(2) Mohan's stream functions are found to be more reliable than those of Nakano and Tien (1968).

(3) The application of the short-range diffusion equation presented by Baird and Hamielec (1962) is shown to be useful not only at very low values of X , but also for values of X as high as about 3, if the Peclet numbers are larger than about 10^4 .

The following may be concluded for mass transfer inside the Newtonian fluid sphere based upon the application of the Mohan stream functions.

(1) The total amount of mass transfer in the dispersed phase, \bar{A}_{mt} , increases slightly with decreases in the continuous phase flow behavior index, when all other variables are held constant. This suggests that the degree of pseudoplas-

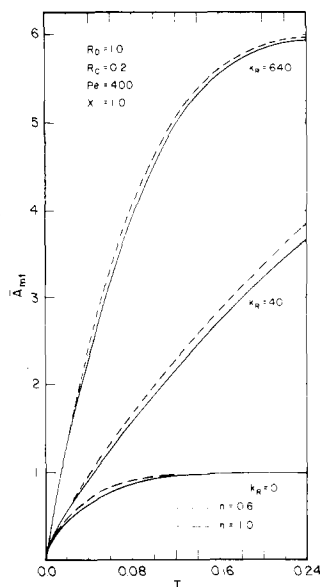


Figure 5. Total mass transferred \bar{A}_{mt} as a function of dimensionless time T for parametric values of the continuous phase flow behavior index n and the reaction number k_R .

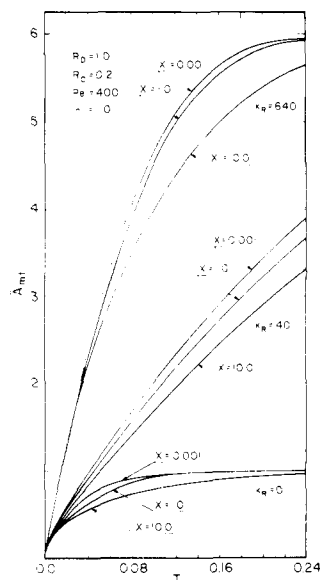


Figure 6. Total mass transferred \bar{A}_{mt} as a function of dimensionless time T for parametric values of the viscosity ratio parameter X and the reaction number k_R .

ticity of the continuous phase does not play an important role in the dispersed phase mass transfer mechanism.

(2) \bar{A}_{mt} increases with decreases in the viscosity ratio parameter, X , when all other variables are held constant. This dependence becomes particularly important when a chemical reaction is accompanying mass transfer in the dispersed phase.

Acknowledgment

Türker Gürkan thanks the Turkish Scientific and Technical Research Council for their support in the form of a NATO Scholarship during this investigation.

Nomenclature

a = spherical radius of dispersed phase, cm
 A_i = coefficients in eq 3a and 3b
 \bar{A}_{mt} = total mass transferred, dimensionless

c_A = concentration of component A, mol/l.
 c_B = concentration of component B, mol/l.
 C_A = concentration of component A, c_A/c_{As} , for the dispersed phase and $c_A/c_{A\infty}$ for the continuous phase, dimensionless
 C_B = concentration of component B, c_B/c_{B0} , dimensionless
 D = diffusivity of solute in the continuous phase, cm^2/sec
 D_A = diffusivity of component A in the dispersed phase, cm^2/sec
 D_B = diffusivity of component B in the dispersed phase, cm^2/sec
 E_i, F_i = coefficients in eq 5a and 5b
 k_c = continuous phase mass transfer coefficient, cm/sec
 k_2 = second-order reaction constant, $\text{l./mol}\cdot\text{sec}$
 k_R = reaction number, $k_2 a^2 c_{B0}/D_A$, dimensionless
 K = consistency index of power-law fluid
 K_1 = coefficient in eq 13a and 13b, $\frac{3}{4} - (A_1/2)(\sigma + 1)$, dimensionless
 n = flow behavior index
 Pe = Peclet number, $2aV_\infty/D_A$, for dispersed phase and $2aV_\infty/D$, for continuous phase, dimensionless
 r = radial distance, cm
 R_C = concentration ratio, $z c_{As}/c_{B0}$, dimensionless
 R_D = diffusivity ratio, D_B/D_A , dimensionless
 Sh = continuous phase Sherwood number, $k_c 2a/D$, dimensionless
 t = contact time, sec
 T = time, $D_A t/a^2$, dimensionless
 U_r = radial velocity, cm/sec
 U_θ = tangential velocity, cm/sec
 V_y = radial velocity, U_r/V_∞ , dimensionless
 V_θ = tangential velocity, U_θ/V_∞ , dimensionless
 V_∞ = relative velocity between sphere and continuous phase fluid, cm/sec
 X = $(\mu_d/K)(a/V_\infty)^{n-1}$, viscosity ratio parameter, dimensionless
 y = radial displacement from center of sphere, r/a , dimensionless
 z = stoichiometric coefficient, $A + zB \rightarrow \text{products}$

Greek Letters

θ = angular displacement from front stagnation point
 μ = viscosity, P
 ρ = density, g/cm^3
 σ = exponent in eq 3a and 3b
 γ = exponent in eq 5a and 5b
 τ_j^i = stress tensor

Δ_j^i = rate of deformation tensor
 Δy = radial increment in finite difference equations
 $\Delta\theta$ = angular increment in finite difference equations

Subscripts

A = component A
 B = component B
 c = continuous phase
 d = dispersed phase
 M = Mohan
 NT = Nakano and Tien
 0 = initial
 s = surface
 ∞ = infinity

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Received for review May 2, 1975

Accepted October 14, 1975

Supplementary Material Available: Tables III, IV, and V, containing data for the mass transfer of bubble flow through fluids (nine pages). Ordering information is given on any current masthead page.