with

$$F_i = -t_{i0} \quad (A7)$$

Subscript 0 indicates that the value of this partial derivative is taken with either the values of parameters obtained at the end of the previous iteration or the initial values. In the calculation of each D_{ij}, it is assumed that the terms of matrix $\mathcal{VAR}(r_i)$ do not vary. In fact, they are parameter dependent and they are recalculated at each step; however, this dependence is small near the optimum and the procedure was found always convergent for all the examples studied.

The F_i's (Equations A6 and A7) are deviation terms which can be considered as random variables generated from a set of measured variables. If one assumes a normal distribution for t_{i0}, it can be shown that $\sigma^2(F_i)$ is unity and $\text{cov}(F_i, F_j)$ is zero because the errors are independent in different experiments. Therefore, the variance-covariance matrix $\mathcal{VAR}(\mathbf{F_i})$ is the unit matrix when all parameter increments ΔC_k are close to zero. The small variations of parameters θC_k produced by small variations in vector **F** are related by the following equation deduced from (A4) and (A6)

$$\mathcal{D}^t \cdot \mathcal{D} \cdot \boldsymbol{\theta}\mathbf{C} = \mathcal{D}^t \cdot \boldsymbol{\theta}\mathbf{F} \quad (A8)$$

where $\mathcal{D}$ is the matrix of which the elements are the D_{ij}'s

$$\mathcal{A} = \mathcal{D}^t \cdot \mathcal{D} \quad (A9)$$

It results from (A8) and (A9)

$$\mathcal{A} \cdot \boldsymbol{\theta}\mathbf{C} \cdot \boldsymbol{\theta}\mathbf{C}^t \cdot \mathcal{A} = \mathcal{D}^t \cdot \boldsymbol{\theta}\mathbf{F} \cdot \boldsymbol{\theta}\mathbf{F}^t \cdot \mathcal{D} \quad (A10)$$

$$\mathcal{A} \cdot \mathcal{VAR}(\mathbf{C}) \cdot \mathcal{A} = \mathcal{D}^t \cdot \mathcal{VAR}(\mathbf{F}) \cdot \mathcal{D} = \mathcal{A} \quad (A11)$$

and finally

$$\mathcal{VAR}(\mathbf{C}) = \mathcal{A}^{-1} \quad (A12)$$

APPENDIX B

The variance covariance matrix of parameters $\mathcal{VAR}(\mathbf{C})$ is a symmetrical $m.\ m$ matrix, and, therefore, it has m real eigenvalues ω_k $(k = 1, \ldots, m)$ associated with m eigenvectors $\mathbf{V}_k$ (of elements $V_{k1}, V_{k2}, \ldots, V_{km}$).

Let

$$\chi_k = \sum_{j=1}^{m} V_{kj} \cdot C_j \quad (B1)$$

the following equations are deduced

$$\sigma^2(\chi_k) = \mathbf{V}_k^t \cdot \mathcal{VAR}(\mathbf{C}) \cdot \mathbf{V}_k \quad (B2)$$

$$\text{cov}(\chi_k, \chi_j) = \mathbf{V}_k^t \cdot \mathcal{VAR}(\mathbf{C}) \cdot \mathbf{V}_j \quad (B3)$$

$\mathbf{V}_k$ and $\mathbf{V}_j$ being eigenvectors, it gives

$$\mathcal{VAR}(\mathbf{C}) \cdot \mathbf{V}_k = \omega_k \cdot \mathbf{V}_k \quad (B4)$$

$$\mathcal{VAR}(\mathbf{C}) \cdot \mathbf{V}_j = \omega_j \cdot \mathbf{V}_j \quad (B5)$$

from (B2), (B3), (B4), and (B5), it is deduced

$$\sigma^2(\chi_k) = \omega_k \mathbf{V}_k^t \cdot \mathbf{V}_k \quad (B6)$$

$$\text{cov}(\chi_k, \chi_j) = \omega_j \cdot \mathbf{V}_k^t \cdot \mathbf{V}_j \quad (B7)$$

As the vectors $\mathbf{V}_k$ and $\mathbf{V}_j$ (with $k \neq j$) are orthogonal and if it is assumed that they are normalized, the following equations are verified:

$$\sigma^2(\chi_k) = \omega_k \quad (B8)$$

$$\text{cov}(\chi_k, \chi_j) = 0 \quad (B9)$$

These last equations indicate that it is possible to find some combinations of parameters the errors of which are independent. The eigenvectors $\mathbf{V}_k$ give the eigendirections of the confidence hyperellipsoid (Ω):

$$\mathbf{C} \cdot \mathcal{VAR}^{-1}(\mathbf{C}) \cdot \mathbf{C} = \beta^2$$

the center of (Ω) being the optimal point $\mathbf{C}^{\text{opt.}}$ previously calculated.

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Interphase Mass and Heat Transfer in Pulsatile Flow

A study on the effects of flow pulsations on interphase mass or heat transfer has been made. The situation corresponds to fully developed flow in a long conduit with a periodic pressure gradient at amplitudes which cause flow pulsations without flow reversal. It is shown that the basic three-parameter formulation can be reduced to a one-parameter problem in the boundary layer formulation. Solutions are developed over wide ranges of the parameters in both the basic and the boundary layer formulations.

The boundary approach gives accurate results over wide ranges of the parameters. Pulsations cause increases in the time-averaged interphase flux at intermediate values of a composite frequency-space variable. However, at small values of this variable the pulsations cause a decrease in the flux such that the overall space-averaged flux is always decreased.

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SCOPE

For the past twenty years a great deal of research has been directed at the question of whether pulsations in the fluid velocity in heat and mass transfer devices can improve the performance of these devices. Investigations of this problem, both theoretical and experimental, have led to numerous contradictions, and there is still confusion over the important basic question as to the circumstances in which pulsations cause an increase or decrease in the average interphase mass or heat transfer.

Interphase mass and heat transfer are of interest in many areas of engineering, and the effect of flow pulsations on the interphase flux clearly is of interest in each

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of these areas. Possible biomedical applications include membrane oxygenators, dialysis devices of various types, and studies on mass transport in the cardiovascular system. To the chemical engineer, the possibility of improving the performance of an exchanger or reactor in which transport of heat or mass is rate controlling is of great practical importance.

The objective of this study is to determine the effects on the time-averaged interphase flux of superimposing a periodic disturbance (in time) on the steady velocity of a fluid passing through a heat or mass transfer device. A simple model of the transfer device is chosen. Briefly, the model consists of a tube of circular cross section. At the wall of the tube, the concentration (or temperature) is maintained at some value different from the concentration (or temperature) of the fluid entering the tube. The fluid is assumed to have a developed velocity profile and to be flowing in the laminar regime with no back flow of the fluid occurring. Furthermore, the fluid phase is assumed to be an incompressible, Newtonian fluid with constant physical properties.

The equations describing diffusion in pulsatile flow are solved by expanding the concentration (or temperature) in a finite Fourier expansion in time. This expansion is substituted into the governing equations and the coefficients of each harmonic are collected and equated to zero. This process generates a set of simultaneous partial differential equations which are solved by implicit finite difference techniques.

The problem described above has recently been studied theoretically by several investigators (Fagela-Alabastro and Hellums, 1969a, 1969b) and a similar problem has been considered by Davidson and Parker (1971). This work is an extension of the work of these previous investigations and is aimed at obtaining results of more general applicability and accuracy.

CONCLUSIONS AND SIGNIFICANCE

The equations describing trace diffusion of material from the wall of a tube into a Newtonian fluid having constant physical properties and flowing through the tube in the laminar regime with a periodic (in time) disturbance superimposed upon the velocity distribution which would exist under steady flow conditions have been solved numerically to a high degree of accuracy. The solutions are restricted to cases in which the fluid does not reverse its direction of flow.

In the boundary layer approximation to the diffusion equation, it was found that the interphase fluxes could be expressed in terms of one parameter, a generalized amplitude parameter, rather than three parameters of the more general formulation. This one-parameter model is very effective in correlating and simplifying the representation of much of the prior work in this field; and it can serve as the basis for simplifying and correlating experimental results. Both the boundary layer solutions and the solutions to the more general equations indicate that the time- and- space-averaged interphase flux for pulsatile flow in a rigid tube is always less than the space-averaged flux for steady flow (both flows having the same mean flow rate). For small enough lengths of tube there is always a decrease in the flux due to pulsations; even though there are local increases in the flux at intermediate lengths, the time- and- space average is always decreased by pulsations. These results indicate that it would not be possible to enhance the performance of, say, a heat exchanger by periodically disturbing the flow on the tube side at least in the ranges of parameters studied.

The effect of flow pulsations on rates of interphase transfer of mass or heat has been studied by several workers in recent years. The prior work can be divided into two categories: (1) external flows along various surfaces such as flat plates, cylinders and spheres, and (2) flow in conduits. The present work is concerned primarily with flow in conduits although of course boundary layer problems in diffusion can often be applied in more general circumstances. The field of interest is further restricted in considering prior work to those studies which have been carried far enough to make quantitative determination of the effect of pulsations on the time-averaged interface mass or heat flux.

There is surprisingly little literature on this restricted class of problems. Reviews of both the theoretical and experimental work are given by Davidson and Parker (1971) and by Fagela-Alabastro and Hellums (1969). Only a few studies will be discussed herein to illustrate the sometimes contradictory nature of the findings. In each case comparisons between pulsatile flow and steady flow are understood to be made at the same average flow rate.

Experiments conducted by Darling (1959) showed that in laminar-pulsatile flow in pipes the space-averaged interphase heat flux is virtually the same as the steady state heat flux. He found that the manner in which the fluid velocity was disturbed from the steady velocity made little difference in the results.

Martinelli et al. (1949), using semisinusoidal velocity disturbances on the tube side fluid of a concentric tube heat exchanger, found that in laminar flow the overall heat transfer coefficient was increased over the steady state coefficient by as much as 30% and that in turbulent flow there was no appreciable change.

West and Taylor (1952), using a reciprocating pump to circulate water through the tube side of a heat exchanger, found a maximum increase of 70% in the overall heat transfer coefficient in turbulent pulsatile flow, whereas no increases were reported in laminar flow.

Edwards, Nellist, and Wilkinson (1973) studied heat transfer in laminar flow in pipes and found that pulsations had no effect on the overall heat transfer, regardless of the amplitude and frequency in Newtonian fluids. In non-Newtonian fluids they observed small (up to 12%) increases.

Krasuk and Smith (1963), in an experimental study of laminar-pulsatile flow mass transfer in pipes, found increases of up to 70% in the mass flux over the steady state flux.

Apparently the first theoretical study was made by Romie (1956). He considered the constant heat flux boundary condition case for flow in a tube and showed that in the limiting case of very large distances downstreams the heat transfer coefficient was less for pulsatile flow than for steady flow.

Davidson and Parker (1971) reported a theoretical study on heat transfer for flow between parallel plates with the isothermal surface boundary condition. They integrated the diffusion equation over the volume of the system and concluded for the limiting case of large distances downstream that the flux must approach that for the steady flow case. This finding does not exclude substantial increases or decreases for finite lengths. Davidson and Parker used a Fourier series expansion of the temperature in which the interaction between harmonics was assumed negligible except for that between the zeroth and first harmonics. The coefficients in the Fourier expansion (and subsequently the heat flux) were obtained by solving the equations for the coefficients using an explicit numerical procedure which required large grid spacings normal to the direction of fluid flow.

Fagela-Alabastro and Hellums (1969) studied the interphase transport in rigid and distensible tubes with the constant concentration (or temperature) wall condition. Boundary layer approximations to the diffusion equation were considered and the concentration (or temperature) was expanded in a power series in the amplitude parameter up to the second-order term. They developed analytical solutions corresponding to the two limiting cases of small and large frequencies and several numerical solutions for the intermediate frequencies. There were a number of interesting results from this work which will be discussed briefly here since the present work may be regarded as a continuation thereof. It was found that the local flux could be either decreased or increased by pulsations depending on the frequency. The increase in flux goes through a maximum and is significant for only a narrow range of frequencies. These increases in the interphase flux are frequency dependent and may become negative for low frequencies in the same way as in the rigid tube case.

The objective of the present work is to develop solutions of more general applicability and accuracy than have been developed by previous investigators. The results have three characteristics different from the prior work. First, the solutions give relatively complete information on the effect of pulsations on the space- and-time-averaged flux, a quantity of obvious practical importance for which only limited results were previously available. Secondly, the solutions are free of the formal limitation to small amplitudes implied in the perturbation procedure previously used. However, the solutions are still formally restricted to the class of flows in which there is no reversal in the direction of flow. Thirdly, the use of the boundary layer simplifications has been examined carefully, and more general equations of change have been applied. It was possible to show that the parametric description of the boundary layer problem is simpler than previously reported.

In the discussion to follow attention will be restricted to Fickian diffusion with constant physical properties. Of course these results can also be designated as representing heat transfer in the usual way. The principal results are for flow in tubes; however, as will be explained below, the boundary layer results can be applied to certain other flows by re-interpreting the meaning of two parameters. The results are for laminar flow and, hence, restricted to Reynolds numbers less than 2000 where laminar flow is stable with respect to pressure pulsations.

In the following sections, the formulation and simplification are given for the boundary layer problem followed by that for the more general case in cylindrical coordinates. Then the method of expansion and solution used in both formulations will be discussed in outline.

FORMULATION OF THE BOUNDARY LAYER PROBLEM

In the diffusion entrance region for a fully developed flow, it is possible to use the thin boundary layer approximations. The approximation and the region of validity are discussed in more detail by Fagela-Alabastro and Hellums (1969a) and by McMichael (1972).

The velocity distribution in response to a periodic axial pressure gradient in a long rigid tube has been known since the early work of Sexl (1930):

$$V_z^* = \left(1-\left(\frac{r}{R}\right)^2\right) - Re\left\{\frac{4i\lambda}{\omega^2}\left[1-\frac{J_0\left(\omega\frac{r}{R}i^{3/2}\right)}{J_0(\omega i^{3/2})}\right]e^{i\beta t}\right\} \quad (1)$$

where the pressure gradient is of the form

$$\frac{\partial P}{\partial z} = \left(\frac{\partial P}{\partial z}\right)_0 (1+\lambda e^{i\beta t}) \quad (2)$$

The velocity distribution near the tube wall (that is, small values of y) can be obtained from Equation (1) by use of Taylors series through the first-order term in y:

$$V_z^* = 2\frac{y}{R} + Re\left\{\frac{4i^{5/2}\lambda y}{R\omega}\,\frac{J_1(\omega i^{3/2})}{J_0(\omega i^{3/2})}e^{it^*}\right\} \quad (3)$$

This expression can be written in a much more compact form as

$$V_z^* = 2\frac{y}{R}(1+\Lambda\cos(t^*+\Omega)) \quad (4)$$

where

$$\Lambda = \frac{2\lambda M_1(\omega)}{\omega M_0(\omega)} \quad (5)$$

and

$$\Omega = \theta_1(\omega) - \theta_0(\omega) - \frac{3\pi}{4} \quad (6)$$

and $M_k(\omega)$ and $\theta_k(\omega)$ are the modulus and argument of $J_k(\omega i^{3/2})$.

The function Λ/λ can be evaluated once and for all as a function of the frequency parameter, ω (see Figure 1) and hence the amplitude of the pressure fluctuation λ will not appear directly in the differential equations describing the problem. In fact, the development of Equation (4) and the introduction of Λ, a generalized amplitude parameter, makes it possible to eliminate both λ and ω as parameters in the final formulation.

The boundary layer diffusion equation is given by

$$\frac{\partial\Psi}{\partial t} + V_z\frac{\partial\Psi}{\partial z} = \mathcal{D}\frac{\partial^2\Psi}{\partial y^2} \quad (7)$$

with conditions on Ψ (t, y, z)

$$\begin{aligned} \Psi(t, 0, z) &= 1 \qquad z > 0 \\ \Psi(t, y, 0) &= 0 \qquad y > 0 \\ \Psi(t, \infty, z) &= 0 \qquad z > 0 \end{aligned} \tag{8}$$

In addition, attention is restricted to periodic solutions with period $2\pi/\beta$. Combining Equations (4) and (7) and selecting variables to minimize the number of parameters by use of the Hellums and Churchill (1964) method yields a one-parameter problem:

$$\frac{\partial \Psi}{\partial \tau} + \sigma(1 + \Lambda \cos \tau) \frac{\partial \Psi}{\partial \xi} = \frac{\partial^2 \Psi}{\partial \sigma^2} \tag{9}$$

The boundary conditions are the same as those given in Equation (8) except that τ replaces t, ξ replaces z, and σ replaces y therein.

The independent variables are defined as

$$\tau = t\beta + \Omega, \quad \sigma = \frac{y\omega Sc^{1/2}}{R}$$

$$\text{and} \quad \xi = \left(\frac{z}{2R}\right)\left(\frac{Sc^{1/2}}{Re}\right)\omega^3 \tag{10}$$

Interestingly enough this choice of variables, which reduces the boundary layer case to a problem in one parameter Λ does not seem to have been used by prior workers. Note that frequency is not a parameter in the problem although of course the effect of frequency is implicit through the definitions of τ, ξ, σ, and Λ. McMichael (1972) has shown how the results of several prior workers including Hellums and Alabastro (1969) and Fortuna and Hanratty (1971) can be compressed and simplified by using the variables of Equation (10). Results presented in terms of two parameters are readily shown to be repetitious. Families of curves collapse into a single curve if expressed in terms of the variables given above.

Another change of variable which will place the problem in a form more suitable for solution will now be considered. Notice from the boundary conditions, Equation (8), that Ψ is not defined at the origin and, of course, Ψ and its derivatives cannot be continuous there. Now consider a transformation of independent variables from (τ, ξ, σ) to (τ, ξ, η) where $\eta = \sigma/\xi^{1/3}$. Equation (9) becomes

$$\xi^{2/3} \frac{\partial \Psi}{\partial \tau} + \eta\xi(1 + \Lambda \cos \tau) \frac{\partial \Psi}{\partial \xi} - \frac{\eta^2}{3}(1 + \Lambda \cos \tau) \frac{\partial \Psi}{\partial \eta} = \frac{\partial^2 \Psi}{\partial \eta^2} \tag{11}$$

with conditions

$$\begin{aligned} \Psi(\tau, \xi, 0) &= 1 \qquad \xi \geqq 0 \\ \Psi(\tau, \xi, \infty) &= 0 \qquad \xi \geqq 0 \end{aligned} \tag{12}$$

The conditions for $\xi = 0$ must be considered in more detail. It is assumed that as $\xi \to 0$, the derivatives of Ψ remain bounded. Then as $\xi \to 0$, Ψ satisfies an equation consisting of the last two terms of Equation (11). The solution of this equation yields the desired condition on the dimensionless concentration Ψ at $\xi = 0$

$$\Psi(\tau, 0, \eta) = 1 - \frac{\int_0^{\eta(1+\Lambda \cos \tau)^{1/3}} e^{-x^3/9}\, dx}{\int_0^\infty e^{-x^3/9}\, dx} \tag{13}$$

Equation (13) is recognized as the limiting case of very low frequencies or the quasi steady solution, and it is now recognized as also representing the limiting case of z (or ξ) $\to 0$. It is of course the classical Leveque solution with the steady velocity of the Leveque problem replaced by the instantaneous value of the velocity. Lipkis (1955) gives a discussion of the Leveque solution and a correction term which may be regarded as a second term in an asymptotic expression for $\xi \to 0$.

The change of space variables from (ξ, σ) to (ξ, η) has another important property illustrated in Figure 2. The boundary layer is stretched from a region wherein $\Psi \to 0$ along $\sigma = \delta_0 \xi^{1/3}$ to a region which is approximately rectangular. In principle, ξ_0 is infinite since $\Psi \to 0$ asymptotically, but in numerical calculations a finite δ_0 can be found for which Ψ is approximately zero to any desired degree of accuracy. The rectangular region and the smoothly varying condition along $\xi = 0$ are both highly advantageous in applying numerical methods to the problem.

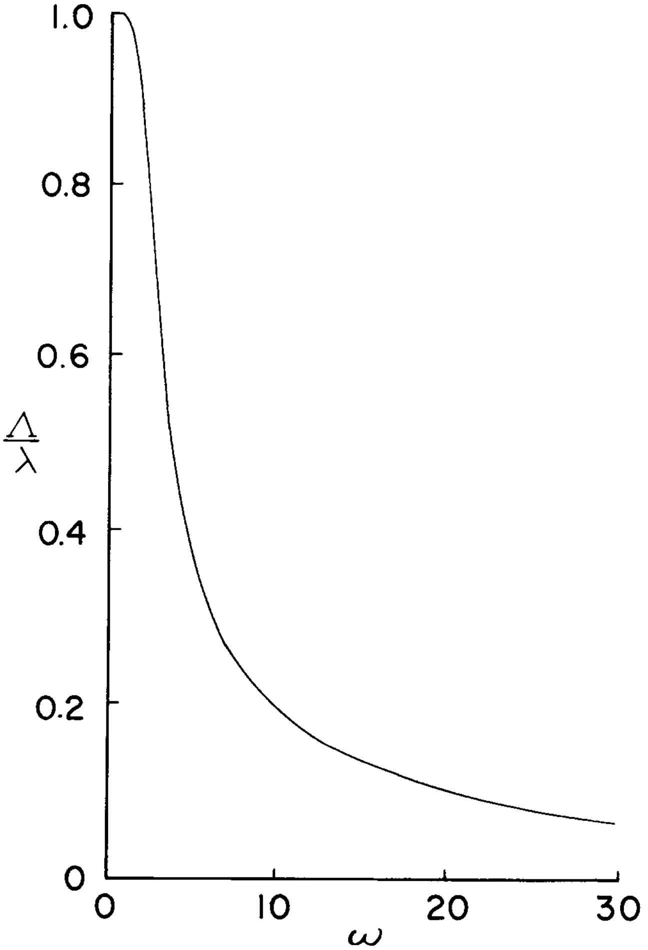

Fig. 1. The generalized amplitude parameter.

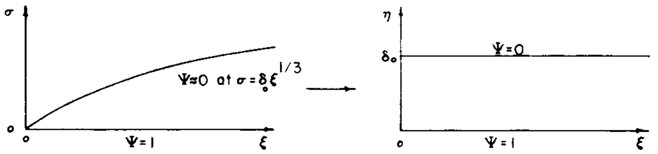

Fig. 2. Stretching of the boundary layer.

FORMULATION IN CYLINDRICAL COORDINATES

The boundary layer approximation in principle is the asymptote for small z and may be of questionable validity for some values of the parameters, especially since the flow pulsations may increase the boundary layer thickness. Therefore, a more general formulation in cylindrical coordinates has been investigated.

The dimensionless equation in cylindrical coordinates neglecting axial diffusion is

$$\omega^2 Sc \frac{\partial \Psi}{\partial \tau} + \frac{\partial}{\partial z^*} (V_z^* \Psi) = \frac{1}{\rho} \frac{\partial}{\partial \rho} \left(\rho \frac{\partial \Psi}{\partial \rho} \right) \quad (14)$$

It can be seen that the parametric description of the problem is much more complicated than the boundary layer case which was reducible to a single parameter Λ. The cylindrical formulation is a three-parameter problem: $\omega^2 Sc$ from Equation (14); and the two independent parameters in the velocity distribution—λ and ω. The velocity distribution is the complete one from Equation (1) rather than the simpler boundary layer approximation of Equation (3) or (4). The generalized boundary layer amplitude parameter Λ has been chosen for use instead of λ because it is known from the boundary layer analysis that to a first approximation ω will not be an independent parameter if such a choice is made. The use of Λ instead of λ is permissible since Λ is determined uniquely by λ and ω through Equation (5) as displayed in Figure 1. Therefore results will be expressed in terms of the three parameters Sc, Λ, and ω.

Equation (14) is in terms of dependent variables (τ, z^*, ρ). For small z^* there is the same motivation to change space variables as in the boundary layer case discussed previously. Hence a transformation is made to the system (τ, z^*, θ) where $\theta = (1 - \rho)/(z^*)^{1/3}$. The transformed equation is to be satisfied with conditions of the form of Equations (12) and (13) with the (ξ, η) space variables replaced by the pair (z^*, θ).

Before going into the numerical method there is another aspect of the differential problem which should be discussed. The use of the second of conditions (12) implies that for all z^*, Ψ approaches the entrance value (zero) to a desired accuracy for some finite value of θ, say θ_{max}. A finite value such as this θ_{max} must be adopted in numerical work. This approach is entirely satisfactory over the range of values of z^* of most practical interest. However, the approach must always fail for sufficiently large z^*. As z^* increases, eventually $(\theta_{max})(z^*)^{1/3}$ approaches unity, corresponding to $\rho = 0$, at which time this approach must be abandoned. Roughly speaking, this condition corresponds to the diffusion boundary layer extending throughout the tube. In the numerical procedure integration is carried out forward in z^*, the time-like variable. The procedure described above was terminated at $z^* = 10^{-3}$ and switched to a large z^* procedure now to be described.

In the large z^* procedure, the original independent space variables (z^*, ρ) were used instead of the (z^*, θ) system discussed above. Since the z^*, ρ system is the basic cylindrical coordinates, no condition at the center line is needed in posing the problem although some authors choose to write $\partial\Psi/\partial\rho = 0$ on $\rho = 0$. For the thick boundary layer (large z^*) region, the (z^*, ρ) system is advantageous. The only difficulty in using the coordinate system is in establishing the initiating conditions at the z^* position where the small z^* procedure is terminated ($z^* = 10^{-3}$). A uniform grid spacing in terms of the θ variable yields a variable spacing when expressed in terms of ρ. Hence, an interpolation procedure had to be used to match the two solutions. Otherwise the Fourier expansion, the equations, and the numerical procedure, while different in detail, are the same in the most important ways. Therefore, only the small z^* case will be discussed here and even that discussion will be restricted to the boundary layer case since our results showed it to be the more important. Complete details of all cases are given by McMichael (1972).

FOURIER EXPANSION AND NUMERICAL PROCEDURE

Equation (11) with conditions given by Equations (12) and (13) and the requirement of periodicity was solved by two independent methods: (1) a sixth-order regular perturbation procedure in Λ, and (2) a Fourier expansion method. In both cases a system of parabolic partial differential equations is generated and solved by a modified Crank-Nicholson numerical method. The perturbation procedure is described by McMichael (1972) and will not be discussed here. Excellent agreement in results was found by the two methods. Remarks will be confined to the Fourier series approach since in principle it is applicable for large amplitudes and since it is the method used in the more general cylindrical case.

The dimensionless concentration Ψ is approximated by the truncated Fourier series:

$$\Psi \approx \sum_{l=0}^{2N} \alpha_l(\xi, \eta)\, V_l(\tau) \quad (15)$$

in which the functions V_l are the usual orthonormal trigonometric functions:

$$V_0(\tau) = \frac{1}{\sqrt{2\pi}}$$

$$V_1(\tau) = \frac{1}{\sqrt{\pi}} \cos\tau \qquad V_2(\tau) = \frac{1}{\sqrt{\pi}} \sin\tau$$

$$\vdots \qquad\qquad \vdots \quad (16)$$

$$V_{2N-1}(\tau) = \frac{1}{\sqrt{\pi}} \cos N\tau \qquad V_{2N}(\tau) = \frac{1}{\sqrt{\pi}} \sin N\tau$$

The Fourier expression is introduced into Equation (11), multiplied by V_j, integrated over a period with respect to τ, and the orthogonality property of the V_j's is used. This procedure yields a system of $2N + 1$ simultaneous, linear, parabolic partial differential equations in the functions $\alpha_l(\xi, \eta)$ in which ξ is the time-like variable:

$$\xi^{2/3} \mathbf{A}\, \boldsymbol{\alpha} + \xi\eta(\mathbf{I} + \Lambda\mathbf{C}) \frac{\partial \boldsymbol{\alpha}}{\partial \xi} - \frac{\eta^2}{3} (\mathbf{I} + \Lambda\mathbf{C}) \frac{\partial \boldsymbol{\alpha}}{\partial \eta} = \frac{\partial^2 \boldsymbol{\alpha}}{\partial \eta^2} \quad (17)$$

where

$\boldsymbol{\alpha}$ is the vector with elements α_l, $l = 0, 1, 2, \ldots, 2N$

$\mathbf{A}$ is the $2N + 1$ square matrix with elements $a_{ij} = \langle V_i, \partial V_j/\partial\tau \rangle$

$\mathbf{C}$ is the $2N + 1$ square matrix with elements $c_{ij} = \langle V_i, V_j \cos\tau \rangle$, and

$\langle x, y \rangle = \int_0^{2\pi} xy\, d\tau$ defines the inner product.

The boundary conditions on the problem for $l = 0, 1, 2, \ldots 2N$ are

$$\alpha_0(\xi, 0) = \sqrt{2\pi}$$

$$\alpha_l(\xi, 0) = 0, \qquad l \neq 0 \quad (18)$$

$$\alpha_l(\xi, \infty) = 0, \qquad \text{for all } l$$

and the initial conditions are simply the Fourier coefficients of $\Psi(\tau, 0, \eta)$ from Equation (13):

$$\alpha_l(0, \eta) = \langle \Psi(\tau, 0, \eta), V_l \rangle \quad (19)$$

To illustrate the form of the problem, it can be said each equation is of the form

$$\xi\eta \frac{\partial \alpha_l}{\partial \xi} = \frac{\partial^2 \alpha_l}{\partial \eta^2} + f_l(\alpha_{l-2}, \alpha_{l-1} \ldots \alpha_{l+2}) \quad (20)$$

where $f_l(\alpha_{l-2}, \alpha_{l-1}, \ldots, \alpha_{l+2})$ denotes all the lower order terms in the equation and illustrates the simultaneous nature of the equations. There is interaction of each α_l (including α_0) with at least one other α_l, and with at most the four α_l with most nearly the same index.

In summary, the original problem has been replaced by Equations (17) subject to conditions given by Equations (18) and (19). The changes of variable and the Fourier expansions have led to a problem of a type well suited for numerical integration: the function and its derivatives in terms of the transformed variables are continuous including the boundary and initial points; the region of integration has been mapped into a semi-infinite rectangle; there is no need for variable grid spacing because the boundary layer has been stretched; and, finally, the Fourier expansion has yielded a system of parabolic equations of the form for which numerical methods are highly developed.

The well-known Crank-Nicholson numerical method was applied to the system of equations. The ξ, η quarter plane is subdivided in the usual way such that $\xi = k\Delta\xi$ and $\eta = j\Delta\eta$. The vector $\boldsymbol{\mu}_j^k$ is defined as the finite difference approximation to $\boldsymbol{\alpha}(k\Delta\xi, j\Delta\eta)$ and the following difference operators are defined:

$$\delta_z \boldsymbol{\mu}_j^k = (\boldsymbol{\mu}_j^{k+1} - \boldsymbol{\mu}_j^k)/(\Delta z^*) \quad (21)$$

$$\delta_\theta \boldsymbol{\mu}_j^k = (\boldsymbol{\mu}_{j+1}^{k+1/2} - \boldsymbol{\mu}_{j-1}^{k+1/2})/(2\Delta\theta) \quad (22)$$

$$\delta_\theta^2 \boldsymbol{\mu}_j^k = (\boldsymbol{\mu}_{j+1}^{k+1/2} - 2\boldsymbol{\mu}_j^{k+1/2} + \boldsymbol{\mu}_{j-1}^{k+1/2})/(\Delta\theta)^2 \quad (23)$$

where $\boldsymbol{\mu}_j^{k+1/2}$ is defined to be $(1/2)(\boldsymbol{\mu}_j^k + \boldsymbol{\mu}_j^{k+1})$. Then the difference equations in $\boldsymbol{\mu}_j^k$ are precisely of the form of Equations (17) in $\alpha(\xi, \eta)$ with the difference operators on the μ variables replacing the corresponding differential operators on the α variables. The α values which are not differentiated are replaced by $\boldsymbol{\mu}_j^{k+1/2}$ the centered value. Similarly, the ξ values are taken at the $k + 1/2$ level. This centering is essential to obtain second-order accuracy in both space variables.

The conditions on the $\boldsymbol{\mu}$ values are the same as those on the $\boldsymbol{\alpha}$ values, Equation (18), where $j = 0$ corresponds to $\eta = 0$ and $j = M$ corresponds to $\eta = \eta_{max}$. Along the line $k = 0$ ($\xi = 0$), either the difference solution or the exact solution to Equation (13) may be used to yield the initial data. The computation procedure consists of computing the values of the $\boldsymbol{\mu}_j^{k+1}$, $j = 1, 2 \ldots M - 1$, from the known values $\boldsymbol{\mu}_j^k$, $j = 1, 2 \ldots M - 1$. The values for $j = 0$ and $j = M$ are the boundary values. The procedure is repeated until any desired $k(\xi)$ level is reached.

The problem at each k level has some interesting and important properties. Suppose the vectors $\boldsymbol{\mu}_j^k$ are ordered to form the vector $\hat{\boldsymbol{\mu}}^k$ where

$$\hat{\boldsymbol{\mu}}^k = \begin{bmatrix} \boldsymbol{\mu}_1^k \\ \boldsymbol{\mu}_2^k \\ \cdot \\ \cdot \\ \cdot \\ \boldsymbol{\mu}_{M-1}^k \end{bmatrix} \qquad \boldsymbol{\mu}_j^k = \begin{bmatrix} \boldsymbol{\mu}_{0,j}^k \\ \boldsymbol{\mu}_{1,j}^k \\ \cdot \\ \cdot \\ \cdot \\ \boldsymbol{\mu}_{2N,j}^k \end{bmatrix} \quad (24)$$

so that in the final elements (those with two subscripts) the first subscript denotes dependent variable number (α number) and the second denotes j position (radial, η or θ position). Then the inversion problem at each stage can be written as

$$\mathbf{D}^{k+1} \hat{\boldsymbol{\mu}}^{k+1} = \mathbf{g}^k \quad (25)$$

In this reordered form, the matrix $\mathbf{D}^{k+1}$ has a very important property in that it is block tridiagonal. That is by no means the same as tridiagonal. Nevertheless, it is possible to invert this matrix directly at each stage by an extension of the method used for tridiagonal matrices. Needless to say, the direct inversion procedure is extremely efficient in comparison to the various iterative schemes often used on this class of problems.

Studies were carried out to check the effects of grid spacing and dimensions and of the number of terms in the Fourier expansion on the computed fluxes. All the results reported here were with five terms in the Fourier expansion ($N = 2$): In test cases the results were the same as those with seven terms ($N = 3$) to at least three significant figures. In the more difficult cylindrical formation other computational parameters were $M = 70$, $\theta_{max} = 8.0$ and Δz^* from 10^{-5} to 10^{-3} depending on the value of z^* and on the Schmidt number. By trial and error the parameters were selected such that the results were independent of the values of M, N, θ_{max}, and Δz^* to three significant figures. McMichael (1972) has presented a detailed discussion of the numerical method including proof of convergence and a discussion of the advantages of using numerically compatible, although slightly inexact, initial data in preference to exact initial data.

RESULTS

All of the results to be discussed here are expressed in terms of time-averaged quantities since these are of most interest in applications. It is interesting to compare both the local and space-averaged flux with the corresponding flux for steady flow at the same mass average velocity. For this purpose the following quantities are defined:

$$\Phi = \Lambda^{-2} \left(\frac{\dfrac{1}{2\pi} \displaystyle\int_0^{2\pi} \left. \frac{\partial \Psi}{\partial \rho} \right|_{\rho=1} d\tau}{\left. \dfrac{\partial \Psi}{\partial \rho} \right|_{\rho=1}^{\Lambda=0}} - 1 \right) \quad (26)$$

$$\overline{\Phi} = \Lambda^{-2} \left(\frac{\dfrac{1}{2\pi z} \displaystyle\int_0^z \int_0^{2\pi} \left. \frac{\partial \Psi}{\partial \rho} \right|_{\rho=1} d\tau dz}{\dfrac{1}{z} \displaystyle\int_0^z \left. \frac{\partial \Psi}{\partial \rho} \right|_{\rho=1}^{\Lambda=0} dz} - 1 \right) \quad (27)$$

The Φ values represent the fractional increase in the interphase flux over the steady state flux divided by Λ^2. Results for various values of Λ show that division by Λ^2 accounts for the Λ dependence fairly accurately: the Φ values so defined depend very little on Λ in the range $(0 < \Lambda < 1)$ studied. In the boundary layer case the flux values for $\Lambda = 0$ (no pulsation) were taken from the well-known analytical solution, Equation (13) with $\Lambda = 0$. In cylindrical coordinates the flux values for $\Lambda = 0$ were developed by solving the equations in the same way as for the nonzero values of Λ. The results for all practical purposes are identical to the well known Graetz-Leveque solution.

RESULTS FOR THE BOUNDARY LAYER APPROXIMATION

Since the boundary layer problem has been reduced to a one-parameter problem, the results can be presented concisely. The results for the local flux are shown in Figure 3 for the complete range of validity of the equations. It can be seen that even the one parameter Λ has relatively little effect in terms of the variables chosen to represent

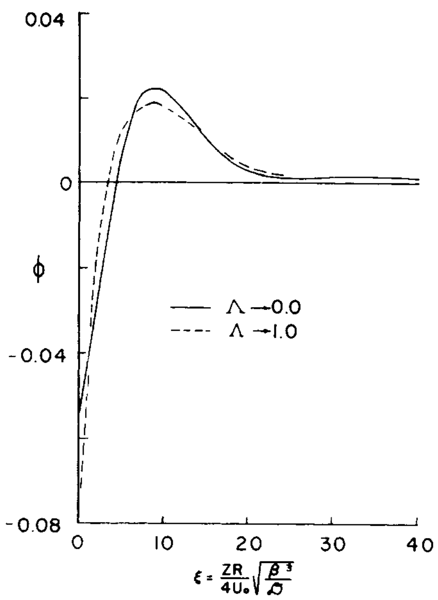

Fig. 3. Effect of pulsations on the time-averaged local interphase flux.

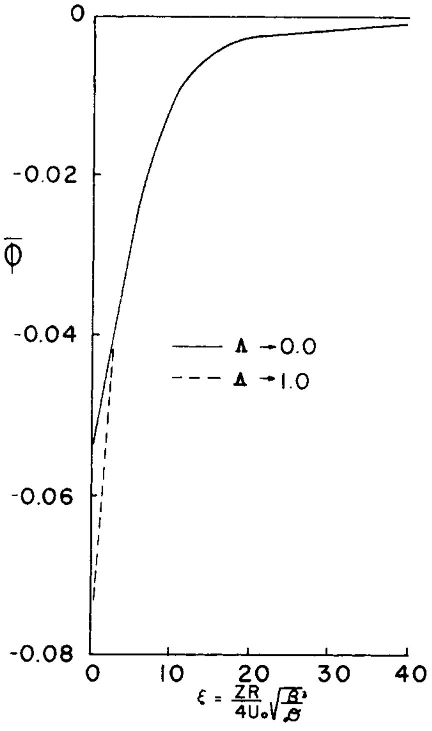

Fig. 4. Effect of pulsations on the time- and space-averaged interphase flux.

the results. The limiting cases of $\Lambda \to 0$ would correspond to either $\lambda \to 0$ or $\omega \to \infty$ (as shown in Figure 1) and is completely equivalent to the steady flow case. Hence, in the limit of $\Lambda \to 0$ the flux increase is zero even though Φ is not. In the limit Φ is of the form zero over zero and the limit is nonzero as shown in the figure.

Notice that for small values of ξ, the flux is actually less in pulsatile flow than in steady flow, for intermediate values there is an increase over a relatively narrow range, then as ξ increases the flux returns to the steady state value. Note that the effect of frequency is slightly more complicated than that of ξ. Changes in frequency affect both ξ and Λ. For example consider increasing frequencies with all other quantities fixed. Beyond the point where Φ is a maximum, the decrease in Φ towards zero is accelerated beyond what one would observe from its effect on ξ because increasing the frequency not only increases ξ but also causes Λ to decrease.

In Figure 3 it is seen that the local flux can either be increased or decreased depending on the axial position and the values of the parameters. A different picture emerges, however, when the space-averaged flux, which is the quantity of primary interest in many applications, is considered. The space-averaged results are shown in Figure 4. For all cases under the boundary layer approach, the net effect of pulsations is to diminish the interphase flux. The negative values of Φ at small ξ more than offset the positive values which occur at intermediate values of ξ. Another point which should be stressed is that the predicted effects are small. Specifically, the greatest magnitude of $\overline{\Phi}$ shown is about 0.08, the decrease at $\xi = 0$. This corresponds to a change due to pulsations of no more than 8%.

Some consideration of the range of validity of the boundary layer approach should be given. Fagela-Alabastro and Hellums (1969a) reported the result that in steady flow the boundary layer approach should be valid over the range $0 < z/R < 0.3\ Re\ Sc$ for $0.1\ Re < L_0/R$, which covers a very wide range of lengths for diffusion in liquids since the Schmidt number for liquids is on the order of 10^3 or higher. The condition $0.1\ Re < L_0/R$ where L_0 is the distance from the inlet of the tube to the inlet of the transfer region guarantees developed flow in the transfer region. The upper limit, $z/R < 0.3\ Re\ Sc$ ($z^* < 0.3$) is due to the thickening of the diffusion boundary layer. In the next section results which show that the boundary layer approach also seems to yield reliable estimates of the flux in pulsatile flow over a wide range of the parameters will be presented.

Before leaving the boundary layer case it should be pointed out that the solutions given here for flow in a tube actually apply to all other boundary layer flows in which the velocity is periodic in time and independent of the space dimension measured along the boundary. The parameters R, Λ, and Ω enter the diffusion problem only through the velocity distribution, Equation (3). Therefore, the solutions apply to any velocity distribution of the form of Equation (3) with appropriate redefinitions of R, Λ, and Ω.

RESULTS IN THE CYLINDRICAL FORMULATION

It is interesting to consider the cylindrical case and to compare the results to those from boundary layer theory. The local and average values of the flux variable are shown in Figures 5 and 6, respectively, as functions of the space variable z^*. The shape of the curves is much the same as in the boundary layer case except that a logarithmic

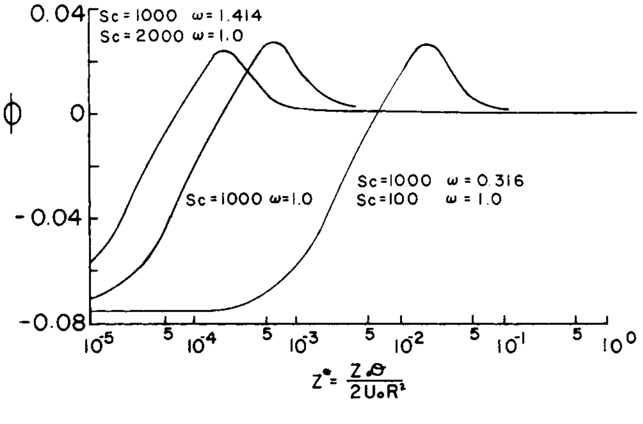

Fig. 5. The time-averaged local flux for the cylindrical system.

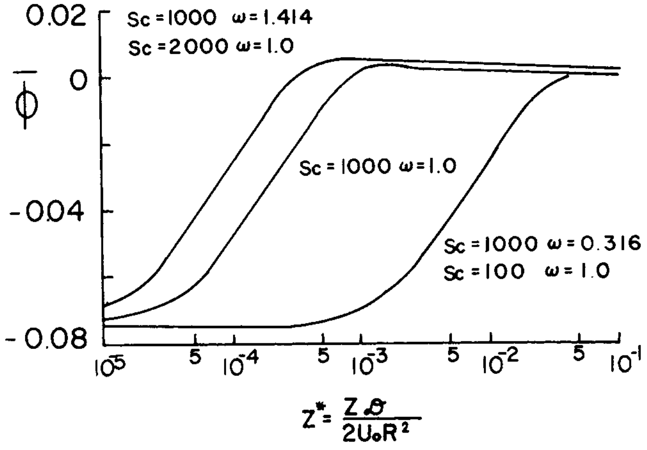

Fig. 6. The time-and space-averaged interphase flux for the cylindrical system.

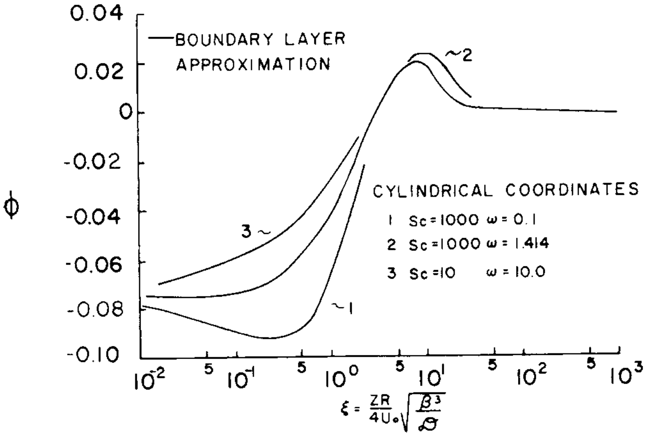

Fig. 7. Comparison of the boundary layer approximation and the cylindrical solution—local flux.

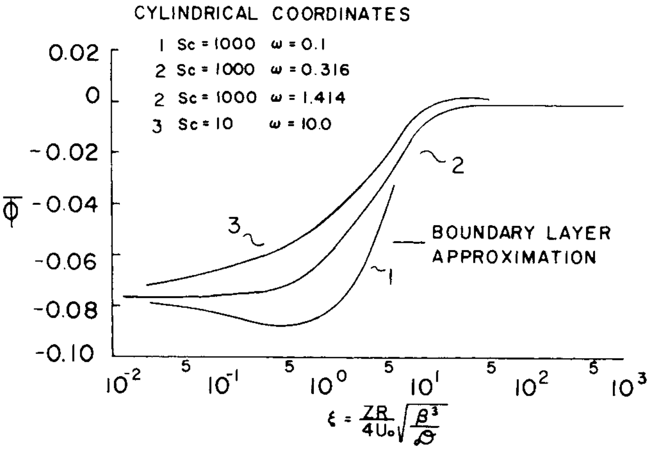

Fig. 8. Comparison of the boundary layer approximation and the cylindrical solution—space-averaged flux.

space variable scale has been introduced so results over a wider range of z^* can be displayed. The curves differ for different values of the parameters Sc and ω, but the important conclusion that the space-and-time averaged flux is always less for pulsatile flow applies as in the boundary layer case. Notice that the upper and lower curves in these figures both have two sets of parameters associated with them. The results were identical within the scale of the figures for the two sets of the parameters. It can be seen that identical results were obtained for different values of the two parameters ω and Sc providing the product $\omega^2 Sc$ was the same. This finding would be predicted from boundary layer theory. It is known that ω is an independent parameter in these more general equations. However, by the choice of variables the effect of ω has been minimized.

The figures show that the flux variables approach zero for large z^*. In some cases the calculations were extended beyond the range shown on the figure, and it was found that Φ increased again for very large z^*. However, the increase in flux found was extremely small and may be an artifact of the numerical procedure. In any event the calculated increase cannot be of consequence because the values of z^* are outside the practical range. They are outside the practical range because the flux, both pulsatile and steady, is many orders of magnitude smaller than the flux in the entrance region. In effect Φ approaches the zero-over-zero form as z^* increases and a significant change in Φ can be caused by insignificant changes in one of the fluxes.

It is interesting to look at these same results expressed in terms of the boundary layer variable ξ as shown in Figures 7 and 8. It is seen that the boundary layer variables do an excellent job of correlating the results together. The deviations in the figures are on an expanded scale and are smaller than would be indicated on superficial observation. In fact, the largest of these deviations from the boundary layer results is about 2%. The largest deviations from boundary layer theory are for the smallest values of Sc, as might be expected. Since the emphasis in this work is on diffusion in liquids, smaller values of the Schmidt number have not been examined in detail, but in such cases boundary layer theory would be less accurate. All the results discussed in this section have been for a fixed value of the parameter Λ. However, other results not reported here showed that, as in the boundary layer case, Λ has little effect on results expressed in terms of Φ and $\overline{\Phi}$.

DISCUSSION

The results presented have all been on the time-averaged interphase flux in the diffusion entrance region. McMichael (1972) presents much more detailed results. He also gives results for additional values of the parameters, and the results of a careful series of studies on the accuracy and convergence of the methods of solution.

The results of Figures 5 through 8 show that the boundary layer approximation is a valid approximation over a somewhat wider range of parameters than might be expected without such detailed study. In setting up the boundary layer problem, it was found that it can be reduced to a single one-parameter problem and that even the one remaining parameter has little effect for practical purposes on an appropriately chosen flux variable. Hence, the results can be presented in an extremely concise way as shown in Figures 3 and 4.

The most important finding (Figures 4, 6, and 8) is that the space-and time-averaged flux is always diminished by flow pulsations. There may be local increases in the flux (Figures 3, 5, and 7), but for all axial positions the space- and time-averaged value is always less than would be obtained in steady flow at the same mass average velocity.

The effects of flow pulsations in rigid tubes are small and are predicted with adequate accuracy by boundary layer theory. In fact, in many applications satisfactory accuracy could be obtained by ignoring the effect of the pulsation. The largest effect determined is a decrease in flux of about 8%, which is smaller than the inherent uncertainty in many mass transfer correlations.

The results presented here are for circumstances for which there is no flow reversal. That is to say, v_z oscillates about some mean value but is positive at all radial positions at all times. Values of the parameters λ and ω for which v_z is always positive are those for which $\Lambda < 1$ as determined using Figure 1. It can be seen from the boundary conditions, Equations (8), that a condition of uniform concentration along $z = 0$ was imposed. This condition could not be imposed in cases for which flow reversal causes upstream transport by convection during some parts of the cycle. Therefore, solutions for $\Lambda > 1$ would require different treatment from that given here. The character of the differential equations is quite different. Intuitive arguments can be advanced to suggest that for $\Lambda > 1$ the effect on the flux would be less than the effect reported here for $\Lambda < 1$. However, such intuitive arguments should be used with caution. The complicated dependence of the flux found in this work would be difficult to explain from intuitive arguments even now that the results are known. Any such qualitative arguments would have to account for changes in sign of the flux variable Φ for various values of ω and z^*. Hence, additional study is needed to extend the work to large amplitudes.

Analysis of the experimental results discussed in the introduction indicates there are still unexplained differences in results under circumstances where large amplitude pulsations are used. Some workers report large increases in interphase flux, and others report none. Some of these apparent contradictions may be due to different experimenters using greatly different values of the parameters. Certainly mass transfer experiments ($Sc \approx 1{,}000$) might be expected to differ from heat transfer experiments ($Pr \approx 10$).

NOTATION

$\mathbf{A}$ = $2N + 1$ by $2N + 1$ matrix with elements a_{ij}; $i, j = 0, 1, \ldots, 2N$
a_{ij} = $\langle V_i, \partial V_j/\partial\tau \rangle$
$\mathbf{C}$ = $2N + 1$ by $2N + 1$ matrix with elements c_{ij}; $i, j = 0, 1, \ldots, 2N$
c_{ij} = $\langle V_i, V_j \cos\tau \rangle$
$\mathbf{D}^{k+1}$ = a block tridiagonal matrix defined by Equation (25)
$\mathcal{D}$ = molecular diffusivity, cm²/s
f_l = a function of $\alpha_{l-2}, \alpha_{l-1}, \ldots, \alpha_{l+2}$ defined by Equation (20)
$\mathbf{g}^k$ = a vector of functions evaluated at the kth stage defined by Equation (25)
$J_k(x)$ = Bessel function of the first kind and kth order
M = number of grid spacings in the θ or η direction
$M_k(x)$ = modulus of $J_k(xi^{3/2})$
N = number of harmonics in the Fourier expansion
Sc = Schmidt number, $\nu/\mathcal{D}$
Re = Reynolds number based on $2R$ and U_0
R = radius of the tube, cm
r = radial position, cm
$\mathbf{S}$ = $2N + 1$ by $2N + 1$ matrix with elements s_{ij}; $i, j = 0, 1, \ldots, 2N$
s_{ij} = $\langle V_i \sin\tau, V_j \rangle$
t = time, s
t^* = dimensionless time, βt
U_0 = time-averaged mean velocity in the axial direction, cm/s
$V(\tau)$ = trigometric function of τ defined by Equation (16)
V_z = velocity in the axial direction, cm/s
V_z^* = $V_z/2U_0$ dimensionless axial velocity
y = distance measured from and normal to the tube wall, cm
z = axial distance measured from the entrance of the transfer region, cm
z^* = $(z\mathcal{D})/(2U_0R^2)$

Greek Letters

$\boldsymbol{\alpha}$ = $[\alpha_0, \alpha_1, \ldots, \alpha_{2N}]^T$
α_i = coefficient of $V_i(\tau)$ in the expansion for Ψ
β = circular frequency of pulsation, s⁻¹
δ_0 = value of η at which the boundary layer ends and $\Psi \approx 0$
$(\partial P/\partial z)_0$ = time-averaged pressure gradient in the axial direction, dynes/cm³
η = similarity variable, $\sigma/\xi^{1/3}$
θ = similarity variable, $(1 - \rho)/(z^*)^{1/3}$
θ_k = phase angle of $J_k(\omega i^{3/2})$
θ_{max} = maximum value of θ, at which $\Psi \cong 0$
Λ = generalized amplitude of pulsation, $2\lambda M_1(\omega)/(\omega M_0(\omega))$
λ = dimensionless amplitude of pressure pulsation
$\hat{\boldsymbol{\mu}}^k$ = vector defined by Equation (24) with elements $\boldsymbol{\mu}_j^k$
$\boldsymbol{\mu}_j^k$ = vector defined by Equation (24) with elements $\mu_{l,j}^k$
$\boldsymbol{\mu}_j^{k+1/2}$ = $(1/2)(\boldsymbol{\mu}_j^{k+1} + \boldsymbol{\mu}_j^k)$
$\mu_{l,j}^k$ = finite difference approximation of $\alpha_l(k\Delta z^*, j\Delta\theta)$
ν = kinematic viscosity, cm²/s
ξ = dimensionless axial distance $zR/4U_0\sqrt{\beta^3}/\mathcal{D}$
ρ = dimensionless radial distance, r/R
σ = dimensionless distance from the tube wall, $y\omega Sc^{1/3}/R$
τ = dimensionless time, $\beta t + \Omega$
Φ = ratio of the time-average interphase flux for pulsatile flow to the steady state flux; defined by Equation (26)
$\overline{\Phi}$ = ratio of the space- and time-averaged in interphase flux for pulsatile flow to the space-averaged steady state flux; defined by Equation (27)
Ψ = dimensionless concentration (or temperature), defined to be zero at the entrance and unity at the boundary
Ω = $\theta_1(\omega) - \theta_0(\omega) - 3\pi/4$
ω = frequency parameter $R(\beta/\nu)^{1/2}$
δ_z = difference operator defined by Equation (21)
δ_θ = difference operator defined by Equation (22)
δ_θ^2 = difference operator defined by Equation (23)
$\langle x, y \rangle$ = inner product, $\int_0^{2\pi} xy\,d\tau$

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Diffusion and Reaction in Turbulent Flow of a Power-Law Fluid in a Circular Tube

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Simultaneous diffusion and homogeneous reaction during the turbulent flow of a power-law fluid in a tubular reactor has been studied using the correlations for the velocity profile and eddy diffusivity proposed by Krantz and Wasan. The results of the theory indicate that while the cup-mixing concentration is unaffected by variations in the Reynolds number, the Schmidt number, or the flow behavior index, it is significantly influenced by the reaction parameter and the reaction order.

SCOPE

The flow of a non-Newtonian fluid in various geometries is a problem that has received considerable attention in recent years. For the turbulent flow of a power-law fluid in a pipe, Krantz and Wasan presented correlations for both velocity profile and eddy diffusivity and predicted significant influence of non-Newtonianism on the axial dispersion coefficient. However, the role of non-Newtonianism on chemical reaction in tubular reactors has not been determined so far.

In this work, simultaneous diffusion and homogeneous reaction in a tubular reactor for the turbulent flow of a power-law fluid has been investigated using the velocity profile and eddy diffusivity correlations proposed by Krantz and Wasan. The resulting differential equation has been solved for Neumann boundary conditions using the Crank-Nicholson finite difference sheme.

CONCLUSIONS AND SIGNIFICANCE

The dimensionless cup-mixing concentration is unaffected by variations in the Reynolds number or the flow behavior index since the dimensionless velocity profile for the turbulent flow of a power-law fluid changes by less than 10%, even for wide variations in these parameters. The conversion is insensitive to variations in Schmidt numbers ranging from 10 to 10^6. However, the length of a reactor for a given conversion is inversely proportional to the reaction parameter for all orders of reaction. Furthermore, the cup-mixing concentration is significantly influenced by the reaction order, the conversion being larger for reactions of lower order. It is therefore concluded that for simultaneous diffusion and reaction in turbulent tubular flow of a power law fluid the major factors influencing the conversion are the reaction parameter and reaction order.

The problem of diffusion and reaction in isothermal tubular reactors has received much attention. The laminar flow of Newtonian fluids has been widely studied starting with the work of Lauwerier (1959), Cleland and Wilhelm (1956), and Hsu (1965) for single homogeneous first-order reactions, Vignes and Trambouze (1962) for second-order reactions, Wissler and Schechter (1961) for consecutive first-order reactions and, Walker (1961) and Solomon and Hudson (1967) for simultaneous homogeneous and heterogeneous first-order reactions. The turbulent flow situation, on the other hand, has been neglected. The wall catalyzed first-order reaction was studied by Wissler and Schechter (1962) for the turbulent flow of a Newtonian fluid in a circular tube and by Randhava and Wasan (1972a) for reactions of arbitrary orders. The corresponding problem for homogeneous reactions with Dirichlet boundary conditions was analyzed by Randhava and Wasan (1971, 1972b).

From a practical standpoint, the analogous problem for the flow of non-Newtonian fluids deserves more attention