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Heterogeneous and Homogeneous Reactions in a Tubular Reactor

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Irreversible, first-order, simultaneous heterogeneous and homogeneous reactions in an isothermal tubular reactor under laminar flow conditions are studied. Accurate values of the eigenvalues, eigenfunctions, and radial concentration profiles are found for the dilute system. Criteria are given as to when the homogeneous reaction may be neglected with respect to the heterogeneous reaction and vice versa. It is found that for a certain range of the rate parameters well-known limiting solutions apply. Outside this range, the new solutions must be used.

The interaction of simultaneous, first-order homogeneous and heterogeneous chemical reactions in an isothermal tubular reactor under fully developed laminar flow conditions is studied. The irreversible reaction

$$A \rightarrow \text{products} \qquad (1)$$

is considered at steady state conditions.

A limiting case of this problem, namely, no homogeneous reaction and infinite heterogeneous reaction rate, is the classical Graetz problem for which accurate values of the eigenvalues and vectors were obtained numerically by Brown (*1*) and asymptotically by Sellars et al. (*2*).

Lauwerier (*3*) considered a finite, first-order homogeneous reaction, but no heterogeneous reaction. An asymptotic solution for the large eigenvalues which was independent of the reaction rate constant was deduced. Recently, Hsu (*4*) solved the same problem by numerical integration for one value of the homogeneous rate constant. Cleland and Wilhelm (*5*) also considered the case of no heterogeneous reaction with a finite-difference method and supported their theoretical profiles with experimental data. Katz (*6*) considered heterogeneous reaction alone from an integral equation viewpoint.

Work on combined first-order heterogeneous and homogeneous reactions has been reported by Walker (*7*), who considered both radial and axial diffusion but reported only the first eigenvalue and considered the concentration profiles from an asymptotic viewpoint. Deviations from the one-dimensional case were of prime consideration. Krongelb and Strandberg (*8*) used finite-difference methods to investigate a second-order homogeneous reaction with first-order heterogeneous kinetics.

It is desired to obtain radial concentration profiles and to deduce under what conditions the homogeneous reaction may be neglected in favor of the heterogeneous reaction and vice versa. In this way combined reaction phenomena may be described in terms of one controlling rate parameter.

ANALYSIS

The dimensionless form of the continuity equation for a dilute component is

$$(1-y^2)\frac{\partial c}{\partial x} = \frac{\partial^2 c}{\partial y^2} + \frac{1}{y}\frac{\partial c}{\partial y} - Kc \qquad (2)$$

with boundary conditions

$$\frac{\partial c}{\partial y} = 0 \qquad y = 0 \qquad (3a)$$

$$c = 1 \qquad x = 0 \tag{3b}$$

$$\frac{\partial c}{\partial y} + fc = 0 \qquad y = 1 \tag{3c}$$

where the dimensionless homogeneous rate group K and the heterogeneous rate group f are defined by

$$K = \frac{K'R^2}{D} \tag{4}$$

$$f = \frac{RK''}{D} \tag{5}$$

In formulating Equations (2) and (3) it has been assumed that axial diffusion may be neglected, that fluid properties are constant, and that the solution is dilute so that the Poiseuille velocity distribution is not disturbed.

Equation (2) may be analyzed through a separation of variables approach leading to a solution

$$c = \sum_{n=0}^{\infty} A_n \psi^{(n)}(y)\, e^{-\lambda_n^2 x} \tag{6}$$

where any $\psi^{(n)}$ satisfies the ordinary differential equation

$$\frac{d^2\psi}{dy^2} + \frac{1}{y}\frac{d\psi}{dy} - K\psi + \lambda^2(1-y^2)\psi = 0 \tag{7}$$

Equation (7) is of the Sturm–Liouville form, and the λ are an infinite set of real, positive eigenvalues. The solution functions $\psi(y)$ are the eigenfunctions corresponding to the λ and through Equations (3) are subject to

$$\frac{d\psi}{dy} = 0 \qquad y = 0 \tag{8}$$

$$\frac{d\psi}{dy} + f\psi = 0 \qquad y = 1 \tag{9}$$

Before Equation (7) is solved, a few general conclusions concerning the behavior of λ with variation in the parameters K and f can be made from the theory of eigenvalue equations. First, consider f. If f is increased monotonically from zero to infinity, each eigenvalue will increase monotonically from its value for $f = 0$ to its value for $f = \infty$. Furthermore, the difference $\lambda_{n+1} - \lambda_n$ will be independent of f for sufficiently large n. The behavior with K is similar in that each λ must increase monotonically with monotonically increasing K.

Equation (7) is solved by the Galerkin method (*9*, *10*). The eigenfunctions are expanded in a complete set of trial functions ϕ_i. That is to say

$$\psi = \sum_{i=1}^{N} R_i \phi_i(y) = \sum_{i=1}^{N} R_i \cos \omega_i y \tag{10}$$

where $\phi_i = \cos \omega_i y$ and the R_i are constants.

The functions ϕ_i are a complete set for $N \to \infty$. Therefore Equation (10) is exact for $N \to \infty$. However in practice a finite expansion must be taken. The convergence of the method is tested by comparing results for successive N.

If Equation (10) is substituted into Equation (7) there results

$$\sum_{i=1}^{N} R_i \left\{ \frac{d^2\phi_i}{dy^2} + \frac{1}{y}\frac{d\phi_i}{dy} - K\phi_i + \lambda^2(1-y^2)\phi_i \right\} = \epsilon(y) \tag{11}$$

where $\epsilon(y)$ is the error. By specifying that the error $\epsilon(y)$ is to be orthogonal to the expansion functions $\phi_j(y)$ on the region of integration $y\,dy$ over the interval [0, 1], a set of N equations is obtained; that is

$$\sum_{i=1}^{N} R_i \int_0^1 \left\{ \frac{d^2\phi_i}{dy^2} + \frac{1}{y}\frac{d\phi_i}{dy} - K\phi_i + \lambda^2(1-y^2)\phi_i \right\} \phi_j\, y dy = \int_0^1 \epsilon(y)\ \phi_j(y)\ y dy = 0 \qquad j = 1, \ldots N \tag{12}$$

The trial functions ϕ_i will satisfy the boundary condition Equation (9) if ω_i is a root of the transcendental Equation (13)

$$\omega_i = f \cot \omega_i \tag{13}$$

The boundary condition Equation (8) is identically satisfied.

Equation (12) will possess a nontrivial solution if the secular condition

$$|\,[A] + \lambda^2 [B]\,| = 0 \tag{14}$$

is satisfied. [A] and [B] are matrices whose respective elements α_{ij} and β_{ij} are given by

$$\alpha_{ij} = \int_0^1 \left\{ \frac{d^2\phi_i}{dy^2} + \frac{1}{y}\frac{d\phi_i}{dy} - K\phi_i \right\} \phi_j\, y dy \qquad \begin{matrix} i = 1, \ldots N \\ j = 1, \ldots N \end{matrix} \tag{15}$$

$$\beta_{ij} = \int_0^1 \phi_i\, \phi_j\, (1-y^2)\, y dy \qquad \begin{matrix} i = 1, \ldots N \\ j = 1, \ldots N \end{matrix} \tag{16}$$

The secular equation yields N values of the eigenvalue λ denoted by λ_n, each having an associated set of eigenvectors R_i denoted by $R^{(n)}$ where $n = 1, \ldots N$. The form of Equation (14) may be simplified to the form

$$|\,[\widetilde{A}] + \lambda^2 [I]\,| = 0 \tag{17}$$

where [I] is the unity array, through use of the Gramm–Schmidt orthonormalization procedure (*9*).

Assume

$$\psi = \sum_{i=1}^{N} \widetilde{R}_i M_i(y) \tag{18}$$

where the new expansion functions (linear combination of the old) are given by

$$M_i(y) = \sum_{j=1}^{i} S_{ji}\, \phi_j(y) \qquad i = 1, \ldots N \tag{19}$$

and the S_{ji} are the Gramm–Schmidt orthonormalization coefficients chosen so that

$$\int_0^1 M_i\, M_j\, (1-y^2)\, y dy = \delta_{ij} \tag{20}$$

where δ_{ij} is the Kronecker delta.

The Galerkin equations for the modified trial functions [Equation (18)] which are equivalent to Equation (12) are

$$\sum_{i=1}^{N} \widetilde{R}_i \int_0^1 \left\{ \frac{d^2M_i}{dy^2} + \frac{1}{y}\frac{dM_i}{dy} - KM_i + \lambda^2(1-y^2)M_i \right\} M_j y dy = 0 \qquad j = 1, \ldots N \tag{21}$$

where the error is orthogonal to $M_j(y)$.

With the use of Equation (19) and the orthonormalization condition, the secular equation is

$$|\,[S]^T[A][S] + \lambda^2[I]\,| = 0 \tag{22}$$

where $[\widetilde{A}] = [S]^T[A][S]$, and $[S]$ is the matrix of the S_{ji} and $[S]^T$ is its transpose. The matrix $[S]$ is upper triangular. The eigenvectors of Equation (22), $\widetilde{R}^{(n)}$, are related to the $R^{(n)}$ by

$$[R] = [S][\widetilde{R}] \tag{23}$$

where $[R]$, $[\widetilde{R}]$ are the matrices whose columns are the eigenvectors $R^{(n)}$, $\widetilde{R}^{(n)}$. The R_i are more convenient to use. It should be noted that the eigenvectors and thus the eigenfunctions are not independent. There is one degree of freedom which is removed by setting $\psi = 1.0$ at $y = 0$, that is, normalization at the center line of the reactor.

The coefficients in Equation (6) are found by applying Equation (3*b*)

$$A_n = \frac{\int_o^1 \psi_n\, y(1-y^2)\; dy}{\int_o^1 \psi_n^2\, y(1-y^2)\, dy} \tag{24}$$

with the condition that the ψ_n form an orthogonal set with weight $y(1 - y^2)$ over the interval $[0, 1]$.

The bulk or cup mixing concentration of A is denoted by $\hat{c}_A$, where

$$\hat{c}_A = \frac{\int_o^{2\pi}\int_o^R V_z(r)\, c_A(r, z)\; r dr d\theta}{\int_o^{2\pi}\int_o^R V_z(r)\; r dr d\theta} \tag{25}$$

In dimensionless terms, and after simplification

$$\hat{c} = 4 \sum_{n=1}^{N} A_n\, e^{-\lambda_n^2 x} \int_o^1 \psi_n\, y(1-y^2) dy \tag{26}$$

Computations were carried out in double precision form on an IBM-7094 digital computer. Fifty-five cases were analyzed. These were combinations of the homogeneous parameter $K = 0, 0.001, 0.01, 0.1, 1.0, 10.0, 100.0$, with the heterogeneous parameter $f = 0, 0.001, 0.01, 0.1, 1.0, 10, 100, \infty$. The Schmidt orthonormalization coefficients were calculated by programmed methods as a first step in the analysis. Exact analytical expressions were derived for the integrals arising in the orthonormalization and all subsequent steps in the solution. These expressions were evaluated for different values of the parameters ω_i, f, K, on the IBM-7094. It is to be emphasized that no numerical integrations were carried out.

Only a few solutions of the transcendental Equation (13) are available in the literature (*11*, *12*). The first twenty roots of this equation for the above values of f were computed by the Newton-Raphson method. The eigenvalues and eigenvectors for the secular equation were determined by means of a noniterative method for symmetric matrices due to Householder, Ortega and Wilkinson (*13*). The matrix in question is symmetric because the operator L

$$L = \frac{1}{y}\frac{d}{dy} y \frac{d}{dy} \tag{27}$$

is self-adjoint subject to the boundary conditions given by Equations (8) and (9).

Sixteen term expansions were used in the solutions of the differential equation. The results of this trial were checked against those for a nineteen term expansion. In all cases of combined f and K at least six eigenvalues were found with uncertainty in only the sixth significant figure. The first five eigenfunctions can be reported to five significant figures for the first two and thence to four significant figures. The expansion coefficients are computed from the values of the eigenfunctions, and thence concentration profiles and bulk concentration values are found.

DISCUSSION

Eigenvalues for several cases are given in Table 1.* As noted above the eigenvalues increase monotonically with increasing homogeneous and heterogeneous reaction rate groups. The eigenvalues, eigenfunctions, and concentration profiles agree well with the results of Brown (*1*) and Hsu (*4*) for the cases $K = 0$, $f = \infty$, and $K = 10$, $f = 0$, respectively.

Consider the effect on the eigenvalues of varying the homogeneous rate group K while holding the heterogeneous rate group f constant. For no heterogeneous reaction Lauwerier (3) noted that the value of the homogeneous rate group influenced the lower eigenvalues but had negligible effect on the higher eigenvalues. It can be seen from Table 1 that an analogous statement can be made for $f \neq 0$. Varying K at fixed f influences all the eigenvalues for $K > 1$, while the changes occur primarily in the first eigenvalue for $K \leq 1$. The effect of varying K on the first eigenvalue is the greatest at small values of f.

Consider now the effect of varying f at fixed K. For large K with $f \leq 1$, the effect of varying f is felt to a slight extent and in fact for $K \geq 10$, the influence of f (and thus the heterogeneous reaction) is negligible. For $K \leq 1$ and $f \leq 1$, a change in f changes only the first eigenvalue, and all other eigenvalues can be approximated by ignoring

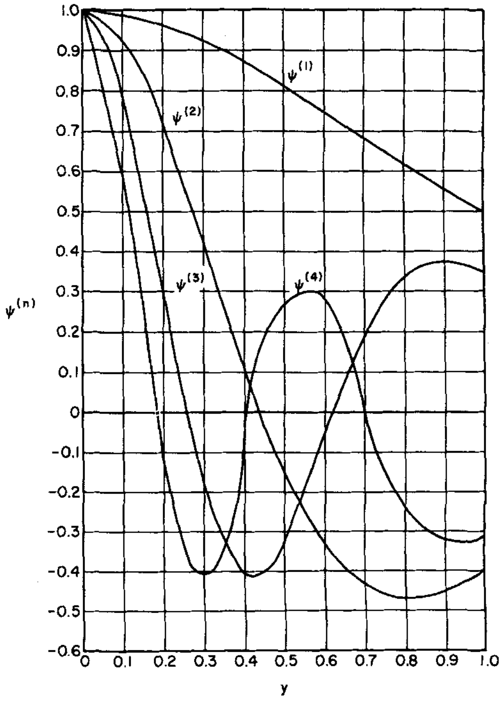

Fig. 1. First four eigenfunctions for $K = 1.0$, $f = 1.0$.

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the heterogeneous reaction. The major changes in the value of the eigenvalues occurs for changes in f between $f = 1.0$ and $f = 10.0$ for fixed K. It can also be stated that for $f \geqq 10.0$, the eigenvalues can be approximated by those found with an infinite wall reaction rate group.

The first eigenfunctions for $K = 1.0$ and several values of f are given in Table 2.* The first four eigenfunctions for $K = 1.0$ and $f = 1.0$ are given in Figure 1. The eigenfunctions for the case of any given $K \neq 0$ and $0 < f < 0.1$ may be approximated by those for $f = 0$; that is, the heterogeneous reaction has a negligible effect on the eigenfunctions. Clear trends in the eigenfunctions are discernable since for a fixed homogeneous rate the eigenfunctions will be nonzero at the wall for a finite heterogeneous rate but are fixed at zero for an infinite wall rate. Thus the eigenfunctions change rapidly in the wall region as the heterogeneous rate increases.

Concentration profiles are determined for a dimensionless position $x = 0.1$. The profiles are all symmetric and have their maximum at the center line as is to be expected. Three selected sets of radial concentration profiles are given in Figures 2, 3, and 4. From Figure 2, and in general for low homogeneous reaction rates ($K \leqq 1$), three distinct regions in the figure are noted. The concentration profiles for $f = 0.1$, 0.01, 0.001 are within 5% of that of $f = 0$, while the concentration profiles for $f = 10$, 100 are close to that of infinite heterogeneous reaction. The region for $0.1 < f < 10$, that is, centered about $f = 1$, is a region where the profiles depend critically on the heterogeneous rate. From Figure 3 it is seen that as the value of the homogeneous rate group increases, the effect of the heterogeneous reaction is lost and concentration values for different f and the same K begin to coincide. The heterogeneous reaction may then be neglected.

By examining the profiles for fixed f and varying K in Figure 4, it is seen that the curves for $K \leqq 0.1$ coincide almost identically with the case of no homogeneous reaction. The concentration profiles differ greatly for different values of the homogeneous rate group in the region between $K = 1.0$ to $K = 10.0$, a much greater difference than, in say, the region $K = 0.1$ to $K = 1$. For large values of the homogeneous rate group, complete conversion occurs.

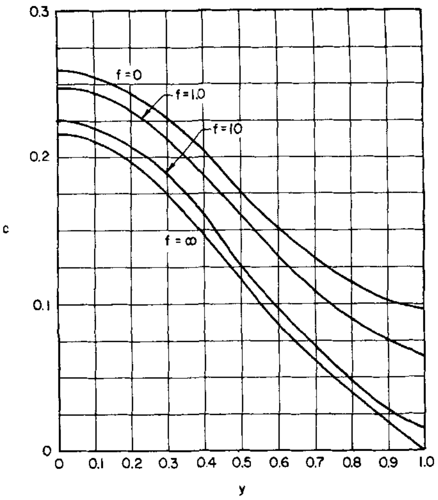

Fig. 3. Radial concentration profiles at $x = 0.1$ for $K = 10.0$ with varying f.

The cup mixing concentrations at dimensionless position $x = 0.1$ as a function of K with f as a parameter are given in Figure 5. The bulk concentration changes by 25% for a given fixed $K \leqq 1.0$ and a change in f from $f = 0.1$ to $f = 1.0$. The corresponding change in the bulk concentration for a change in the heterogeneous reaction group from $f = 1.0$ to $f = 10.0$ is about 40%. Changes outside this range are negligible in comparison. In Figure 6 bulk concentrations for $x = 0.1$ as a function f with K as a parameter are given. A change in the homogeneous

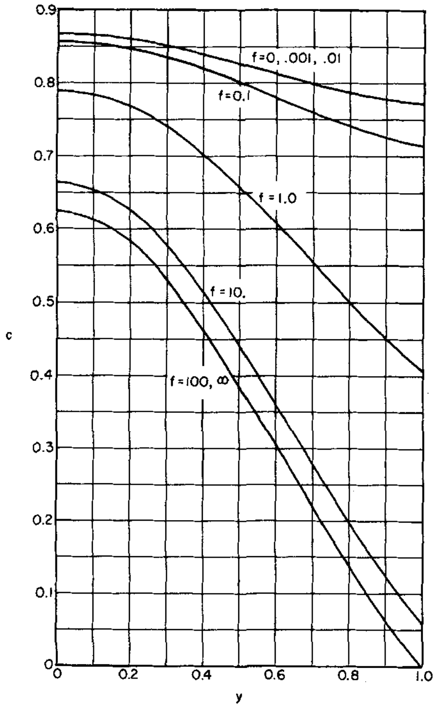

Fig. 2. Radial concentration profiles at $x = 0.1$ for $K = 1.0$ with varying f.

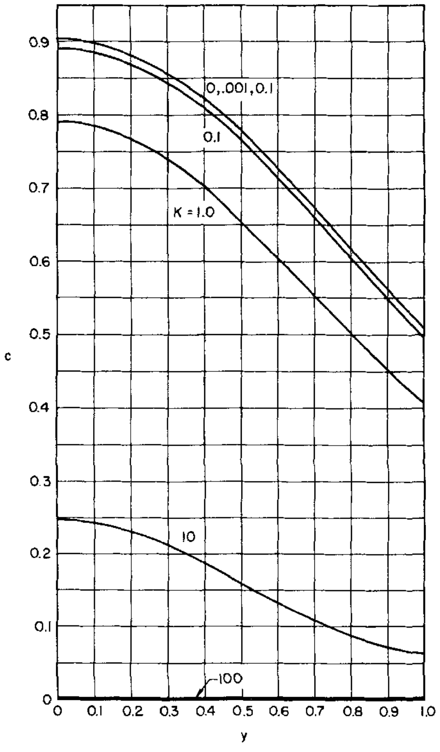

Fig. 4. Radial concentration profiles at $x = 0.1$ for $f = 1.0$ with varying K.

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* See footnote on page 547.

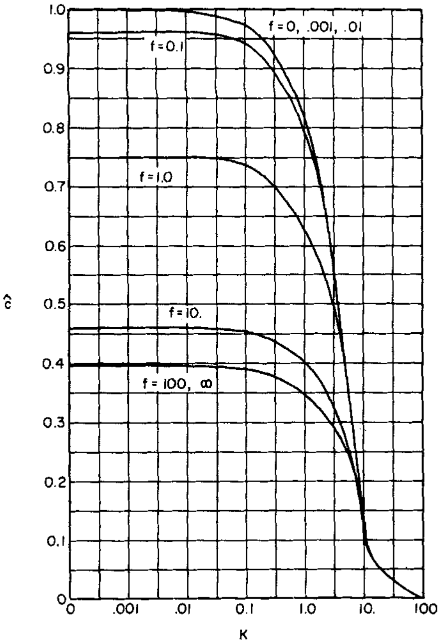

Fig. 5. Bulk concentration dependence on homogeneous rate with heterogeneous rate as a parameter at $x = 0.1$.

rate group from $K = 0.1$ to $K = 1.0$, for a fixed heterogeneous rate group, causes a change in the bulk concentration of 12 to 15%. For a change from $K = 1.0$ to $K = 10.0$, the bulk concentration changes by 70 to 80%.

Although no experimental data are available to test the results, the range of rate parameters covers all possible situations. For gas phase reactions the values of the dimensionless rate constants will be approximately f, $K = 0.1$, while for liquid phase reactions the values will be much higher owing to the lower diffusivities. In fact, quite often the limiting correlations will apply in this phase.

In order to determine the values of K and f for any given gaseous reacting mixture and reactor, the effluent bulk concentration for a system should be experimentally determined. Under conditions of a different diffusivity, due to varying the diluent or pressure, the bulk concentration is remeasured. In order to maintain the same value of the parameter x, the flow rate must be proportionately varied. Reference to Figures 5 and 6 will yield the values of the rate parameters, since changes in the diffusivity produce changes in the bulk concentration dependent on the relative magnitude of the homogeneous and heterogeneous reactions.

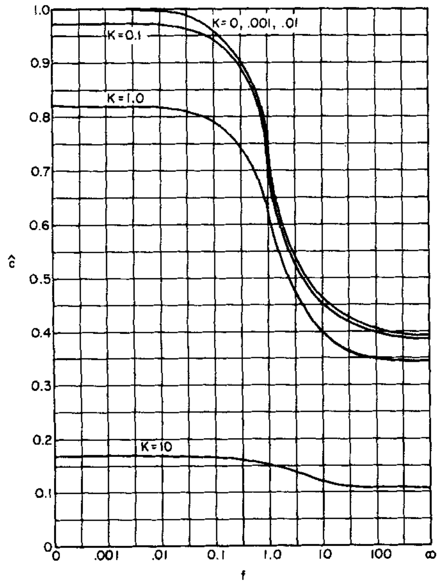

Fig. 6. Bulk concentration dependence on heterogeneous rate with homogeneous rate as a parameter at $x = 0.1$.

CONCLUSIONS

The effect of the heterogeneous reaction on the radial concentration profiles is negligible for low heterogeneous rates, that is, $f \leqq 0.1$ and for large homogeneous rates, that is $K > 10$. Thus the value of the heterogeneous group parameter f can be taken as $f = 0$ and the limiting solutions applied; that is, the homogeneous reaction is the controlling factor. The effect of the homogeneous reaction is to cause percentage changes in the concentration of less than 3% if $K < 0.1$. In this case the heterogeneous reaction rate is controlling. For this range a valid approximation for $f \geqq 10$ is to treat the case as that of infinite wall reaction.

Extremely erroneous results would arise if any limiting case approximations were applied to the cases where both heterogeneous and homogeneous rate groups lie in the range $0.1 < f < 10$, $0.1 < K < 10$. In this region individual concentration profiles must be computed as indicated in this paper. A change in the homogeneous and/or heterogeneous rate have the maximum effect when the rate group changes in the range $f,K = 1.0$ to $f,K = 10$.

ACKNOWLEDGMENT

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NOTATION

A_n = n^{th} concentration expansion coefficient
$[A]$ = matrix of α_{ij}
$[\tilde{A}]$ = matrix as defined by Equation (22)
$[B]$ = matrix of β_{ij}
c = c_A/c_o = concentration of component A, dimensionless
c_A = molar concentration of A
c_o = inlet molar concentration of A
$\hat{c}$ = $\hat{c}_A/c_o$ = bulk concentration, dimensionless
$\hat{c}_A$ = bulk molar concentration of A .
D = diffusivity
f = dimensionless heterogeneous rate parameter as defined by Equation (5)
$[I]$ = identity matrix
K = dimensionless homogeneous rate parameter as defined by Equation (4)
K' = homogeneous rate constant
K'' = heterogeneous rate constant
L = operator as defined by Equation (27)
M_i = i^{th} modified trial function
N = number of terms in series expansion
r = radial coordinate
R = tube radius
$R^{(n)}$ = n^{th} eigenvector in eigenfunction expansion
$\tilde{R}^{(n)}$ = n^{th} eigenvector in modified eigenfunction expansion
R_i = i^{th} element of $R^{(n)}$

$\tilde{R}_i$ = i^{th} element of $\tilde{R}^{(n)}$
$[R]$ = matrix of $R^{(n)}$
$[\tilde{R}]$ = matrix of $\tilde{R}^{(n)}$
S_{ji} = j, i^{th} element of $[S]$
$[S]$ = Gramm–Schmidt orthonormalization matrix
V_z = axial velocity
V = maximum velocity in Poiseuille profile
x = Dz/VR^2 = axial coordinate, dimensionless
y = r/R = radial coordinate, dimensionless
z = axial coordinate

Greek Letters

α_{ij} = i, j^{th} element of $[A]$
β_{ij} = i, j^{th} element of $[B]$
δ_{ij} = Kronecker delta
ϵ = error
λ_n = n^{th} eigenvalue
ϕ_i = i^{th} trial function
ψ_n = n^{th} eigenfunction
ω_i = i^{th} solution of Equation (13)

Superscript

T = transpose

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External Flows of Viscoelastic Materials: Fluid Property Restrictions on the Use of Velocity-Sensitive Probes

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The more pronounced macroscopic features of flows of viscoelastic materials around submerged objects are considered in the light of restrictions imposed on the flow by the Deborah number, a dimensionless group representing a ratio of time scales of the fluid and the flow process. It is seen that one major effect is to thicken the boundary layer appreciably in the region of the leading edge or stagnation point of the object in the fluid; depending on the shape of the object this thickening of the boundary layer may be felt for appreciable distances into the velocity field.

The influence of these and other effects on the use of probes for determination of point values of the velocity of viscoelastic fluids is considered in some detail. The general effect is to impose strong restrictions on the use of such probes and on the interpretation of measurements made with them. These predicted limitations are supported, in a general way, by recent experimental measurements; thus the present macroscopic analysis appears to define several major effects to be considered in subsequent, more detailed investigations.

Some general features of flows external to objects of various geometries and of boundary-layer flows appear to differ grossly between viscoelastic and Newtonian fluids; the purpose of the present paper is to describe these features macroscopically, for ordering purposes, and to make a start on the detailed analysis of the more important of these.

While external flows may be broadly of interest in engineering, the analysis presented is directed particularly toward an understanding of flows around objects inserted in the fluid to sense its local velocity or pressure, such as heated cones or wires, impact tubes, and small bubbles or particles. It will be seen that the deformational behavior

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