Diffusion and Chemical Reaction in a Tubular Reactor with Non-Newtonian Laminar Flow

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The problem treating an irreversible, first-order chemical reaction in an isothermal, tubular reactor operating under steady, non-Newtonian, laminar flow is studied. Numerical results for the eigenvalues, eigenfunctions, expansion coefficients and bulk concentration are reported for Ostwald-de Waele and Prandtl-Eyring fluids. Results show that chemical conversion in a tubular reactor may be significantly influenced by the non-Newtonian characteristics of a fluid.

Much attention has been given to the problem involving an irreversible, first-order chemical reaction in an isothermal, tubular reactor operating under steady, laminar flow. Using a finite-difference numerical technique, Cleland and Wilhelm (*1*) obtained the solution for a Newtonian fluid in which radial molecular diffusion of the reactant was considered. Analytic solutions were determined for the two limiting cases of zero and infinite radial molecular diffusion rates, and experimental data were reported to support their numerical predictions. In addition they reported theoretical conditions for which diffusion may be neglected, which compared well with those given previously by Bosworth (*2*). Lauwerier (*3*) was the first to recognize that a separation of variables was possible and that the resulting characteristic value problem was of the confluent hypergeometric type. Based upon his previous work treating the confluent hypergeometric function (*4* to *6*), Lauwerier obtained an asymptotic solution valid for large eigenvalues. Furthermore, he formulated a power series solution, the numerical work for which was later performed by Wissler and Schechter (*7*) and was found to agree with the results obtained by Cleland and Wilhelm. More recently, Hsu (*8*) solved the same problem by separation of variables and numerical integration of the characteristic value problem for one value of the reaction rate constant. Related problems treating different kinetics (*7*, *9*) and simultaneous heterogeneous and homogeneous chemical reactions (*10* to *12*) in tubular flow have also been studied.

The analogous problem for the flow of a non-Newtonian fluid apparently has not been treated. This problem is of particular concern at this laboratory in the study of thermal pasteurization processing of liquid foods, inasmuch as the death rate of microorganisms is first order and a large number of common liquid foods exhibit non-Newtonian rheological behavior. It therefore seems appropriate to study the tubular flow reaction problem for a non-Newtonian fluid.

THEORY

Non-Newtonian Models

Numerous empirical equations, or models, have been proposed to relate the shear stress τ with the rate of shear dv/dr for non-Newtonian fluids. In this report we shall consider two such models.

Consider the Ostwald-de Waele model (*13*) (also known as the power law model) given by the relation

$$\tau = -\eta \left|\frac{dv}{dr}\right|^{s-1} \frac{dv}{dr} \tag{1}$$

Substitution of Equation (1) into the axisymmetric equations of motion and subsequent integration yield the familiar relation for the velocity profile in a tube of radius R

$$v(r) = \frac{3s+1}{s+1} \langle v \rangle \left[1 - \left(\frac{r}{R}\right)^{\frac{s+1}{s}}\right] \tag{2}$$

which simplifies to the Poiseuille distribution when $s = 1$.

Another model somewhat different from that of Ostwald and de Waele is the Prandtl-Eyring model (*14* to *16*) given by

$$\tau = \mu \operatorname{arcsinh}\left(-\frac{1}{\nu}\frac{dv}{dr}\right) \tag{3}$$

where μ and ν are constants. Unlike the power law model, the latter has some physical significance inasmuch as it can be deduced by kinetic theory (*13*). Substitution of Equation (3) into the equations of motion and integration yields the velocity expression

$$v(r) = \frac{\langle v \rangle}{\omega}\left[1 - \frac{\cosh\left(\frac{r}{R}\,b\right)}{\cosh(b)}\right] \tag{4}$$

where

$$b = \frac{R}{2\mu}\frac{dp}{dz} \tag{5}$$

$$\omega = 1 - \frac{2\tanh(b)}{b} + \frac{2}{b^2} - \frac{2}{b^2\cosh(b)} \tag{6}$$

It is apparent from Equation (3) that the Prandtl-Eyring model predicts pseudoplastic behavior at finite τ and reduces asymptotically to Newton's law of viscosity for $\tau \ll \mu$ with viscosity $\eta = \mu/\nu$.

In order to simplify the development to follow it is convenient to generalize the velocity expressions (2) and (4) by writing them as

$$v(r) = \beta \langle v \rangle \left[1 - \phi\left(\frac{r}{R}\right)\right] \tag{7}$$

where β is a constant and $\phi(r/R)$ is a general dimensionless radial function given by Equations (2) and (4) for the Ostwald-de Waele and Prandtl-Eyring models, respectively.

Analytical Development

The dimensionless form of the steady state continuity equation with first-order homogeneous chemical reaction and negligible axial diffusion of reactant is given by the differential equation

$$\kappa\left(\frac{\partial^2 C}{\partial \xi^2} + \frac{1}{\xi}\frac{\partial C}{\partial \xi}\right) - \beta[1 - \phi(\xi)]\frac{\partial C}{\partial \zeta} - C = 0 \tag{8}$$

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subject to the boundary conditions

$$C = 1 \text{ at } \zeta = 0, \quad 0 \le \xi \le 1 \tag{9a}$$

$$\frac{\partial C}{\partial \xi} = 0 \text{ at } \xi = 0, \quad \zeta > 0 \tag{9b}$$

$$\frac{\partial C}{\partial \xi} = 0 \text{ at } \xi = 1, \quad \zeta > 0 \tag{9c}$$

Here the generalized velocity expression (7) has been used along with the following dimensionless groups

$$C = \frac{c}{c_0} \qquad \xi = \frac{r}{R}$$

$$\kappa = \frac{D}{kR^2} \qquad \zeta = \frac{kz}{\langle v \rangle}$$

The parameter ζ, which may be thought of as a dimensionless axial length, is known as the Damköhler Group I, while κ, which may be considered as a dimensionless molecular diffusivity, is the inverse of the Damköhler Group II.

Equation (8) may be solved by the method of separation of variables to yield the solution

$$C(\xi, \zeta) = \sum_{n=1}^{\infty} a_n \, e^{-\lambda_n^2 \zeta/\beta} \, \Xi_n(\xi) \tag{10}$$

where the Ξ_n satisfy the ordinary differential equation

$$\kappa \left(\Xi_n'' + \frac{1}{\xi} \Xi_n' \right) + \{ \lambda_n^2 [1 - \phi(\xi)] - 1 \} \Xi_n = 0 \tag{11}$$

with boundary conditions

$$\Xi' = 0 \quad \text{at} \quad \xi = 0 \tag{12a}$$

$$\Xi' = 0 \quad \text{at} \quad \xi = 1 \tag{12b}$$

Here the prime marks indicate differentiation with respect to ξ.

Equations (11) and (12) can be seen to comprise a proper Sturm-Liouville system for which there exists an infinite set of real, non-negative eigenvalues λ_n and a corresponding set of real eigenfunctions Ξ_n. The expansion coefficients a_n may be obtained by applying Equation (9a) and the condition of orthogonality of eigenfunctions with respect to the weighting function, $\xi[1 - \phi(\xi)]$.

$$a_n = \frac{\int_0^1 \Xi_n \, \xi [1 - \phi(\xi)] \, d\xi}{\int_0^1 \Xi_n^2 \, \xi [1 - \phi(\xi)] \, d\xi} \tag{13}$$

The method of Galerkin (*17* to *19*) is used to solve Equation (11) by expanding the eigenfunctions in a finite set of N trial functions γ_i

$$\Xi_n(\xi) = \sum_{i=1}^{N} X_{in} \, \gamma_i(\xi) \quad n = 1, \cdots N \tag{14}$$

where the X_{in} are constants and the γ_i are chosen to satisfy Equations (12a) and (12b). Following the method of Singh (*20*), the trial functions γ_i are chosen as solutions of the auxiliary equation, which in this case is the zero-order Bessel's equation. With this choice Equation (14) becomes

$$\Xi_n(\xi) = \sum_{i=1}^{N} X_{in} \, J_0(\alpha_i \xi) \quad n = 1, \cdots N \tag{15}$$

where the α_i are determined from Equation (12b) to be the zeros of the first-order Bessel function. Substituting this finite series, Equation (15), into Equation (11), and according to the method of Galerkin (*10*), by which the error incurred by using this finite representation is required to be orthogonal to each trial function with respect to the weighting function ξ, one can obtain the matrix equation:

$$\mathbf{AX} - \frac{1}{\lambda^2} \mathbf{BX} = 0 \tag{16}$$

Here, $\mathbf{X}$ is the eigenvector corresponding to a given eigenvalue λ, and $\mathbf{A}$ and $\mathbf{B}$ are matrices whose elements are given by

$$A_{ij} = 2 \int_0^1 J_0(\alpha_i \xi) J_0(\alpha_j \xi) \, \xi \phi(\xi) \, d\xi - J_0^2(\alpha_i) \delta_{ij} \tag{17}$$

$$B_{ij} = (\kappa \alpha_i^2 + 1) J_0^2(\alpha_i) \delta_{ij} \quad i, j = 1, \cdots N \tag{18}$$

and δ_{ij} is the Kronecker delta. An important consequence of choosing the approximation functions as solutions of the auxiliary equation has been to make $\mathbf{B}$ diagonal, a fact which will simplify much of the remainder of the problem. Thus the problem of solving the differential Equation (11) has been reduced to one of solving the matrix Equation (16).

In order to solve Equation (16) we seek a transformation matrix $\mathbf{S}$, chosen such that $\mathbf{S}^T\mathbf{BS} = \mathbf{I}$, where $\mathbf{S}^T$ is the transpose of $\mathbf{S}$ and $\mathbf{I}$ is the identity matrix. With this transformation Equation (16) may be written equivalently as

$$\mathbf{H\tilde{X}} - \frac{1}{\lambda^2} \mathbf{I\tilde{X}} = 0 \tag{19}$$

where $\mathbf{H}$ is defined by the relation

$$\mathbf{H} = \mathbf{S}^T\mathbf{AS} \tag{20}$$

The eigenvalues of Equation (19) are the same as those of Equation (16), while the eigenvectors are related by the expression

$$\mathbf{X} = \mathbf{S\tilde{X}} \tag{21}$$

Since $\mathbf{B}$ is diagonal, $\mathbf{S}$ is uniquely given by

$$\mathbf{S} = \mathbf{B}^{-1/2} \tag{22}$$

that is, $\mathbf{S}$ is also diagonal, the elements of which are given by

$$S_{ij} = \frac{1}{\sqrt{\kappa \alpha_i^2 + 1}} \frac{\delta_{ij}}{|J_0(\alpha_i)|} \tag{23}$$

Furthermore, since $\mathbf{A}$ is symmetric and $\mathbf{S}$ is diagonal, it follows from Equation (20) that $\mathbf{H}$ is also symmetric, the elements of which are given directly by

$$H_{ij} = S_{ii} A_{ij} S_{jj} \tag{24}$$

Equation (19) will possess a nontrivial solution if and only if the characteristic equation

$$\left| \mathbf{H} - \frac{1}{\lambda^2} \mathbf{I} \right| = 0 \tag{25}$$

is satisfied. This equation may be solved by standard methods to yield N distinct eigenvalues λ_n as roots of the characteristic polynomial for $\mathbf{H}$ and an associated set of eigenvectors $\tilde{\mathbf{X}}$.

Thus, the solution of Equation (8) is complete and may be summarized as follows. The matrix $\mathbf{H}$ is determined from Equation (24), $\mathbf{A}$ and $\mathbf{S}$ being calculated from Equations (17) and (23), respectively. A set of N eigenvalues and eigenvectors is determined from Equation (25). The eigenvectors are further transformed by Equation (21) to those corresponding to Equation (15), whence N eigenfunctions are finally determined. The dimensionless concentration is then evaluated using Equation (10), truncated at N terms; the expansion coefficients are obtained from Equations (13) and (15).

Of most practical interest is the bulk (or cup mixing) concentration of reactant $\hat{c}$ at any axial position in the tube.

$$\hat{c} = \frac{\int_0^{2\pi}\int_0^R c(r,z)\,v(r)\,r\,dr\,d\theta}{\int_0^{2\pi}\int_0^R v(r)\,r\,dr\,d\theta} \tag{26}$$

Introducing dimensionless variables and substituting Equation (7) and a truncated expansion of Equation (10) into Equation (26) result in

$$\hat{C} = \frac{\sum_{n=0}^{N} a_n e^{-\lambda_n^2 \zeta/\beta} \int_0^1 \Xi_n\,\xi[1-\phi(\xi)]\,d\xi}{\int_0^1 \xi[1-\phi(\xi)]\,d\xi} \tag{27}$$

Introducing Equation (15) for the eigenfunctions and after some simplification, we obtain

$$\hat{C} = \frac{\sum_{n=0}^{N} a_n e^{-\lambda_n^2 \zeta/\beta}\left[\frac{X_{11}}{2} - \sum_{i=1}^{N} X_{in}\int_0^1 J_0(\alpha_i\xi)\,\xi\phi(\xi)\,d\xi\right]}{\frac{1}{2} - \int_0^1 \xi\phi(\xi)\,d\xi} \tag{28}$$

where

$$a_n = \frac{\frac{X_{11}}{2} - \sum_{i=1}^{N} X_{in}\int_0^1 J_0(\alpha_i\xi)\,\xi\phi(\xi)\,d\xi}{\frac{1}{2}\sum_{i=1}^{N}[X_{in}J_0(\alpha_i)]^2 - \sum_{i=1}^{N}\sum_{j=1}^{N} X_{in}X_{jn}\int_0^1 J_0(\alpha_i\xi)J_0(\alpha_j\xi)\,\xi\phi(\xi)\,d\xi} \qquad n = 1, \cdots N \tag{29}$$

Note that the integral obtained in the denominator of Equation (29) is the same as that found in Equation (17).

Ostwald-de Waele Model

Reference to the velocity expression for the Ostwald-de Waele model given by Equation (2) shows that

$$\beta = \frac{3s+1}{s+1}, \qquad \phi(\xi) = \xi^{\frac{s+1}{s}}$$

For the special case of a Newtonian fluid for which $s = 1$, the integrals resulting from substitution of the above terms into Equation (29) may be solved exactly (*20*) to obtain

$$\int_0^1 J_0(\alpha_i\xi)J_0(\alpha_j\xi)\,\xi^3\,d\xi$$

$$= \frac{1}{4}, \qquad i = j = 1 \tag{30}$$

$$= \frac{J_0^2(\alpha_i)}{6}, \qquad i = j \neq 1 \tag{31}$$

$$\doteq \frac{2J_0(\alpha_i)J_0(\alpha_j)(\alpha_i^2+\alpha_j^2)}{(\alpha_i^2-\alpha_j^2)^2}, \qquad i \neq j \tag{32}$$

It should be noted that $\alpha_1 = 0$.

Prandtl-Eyring Model

Similarly, for the Prandtl-Eyring model we have

$$\beta = \frac{1}{\omega}, \qquad \phi(\xi) = \frac{\cosh(b\xi)}{\cosh(b)}$$

Analytical solutions of the resulting integrals in Equation (29) could be found only for the case, $i = j = 1$, for which

$$\int_0^1 \xi\cosh(b\xi)\,d\xi = \frac{1}{b^2}\,[b\sinh(b) - \cosh(b) + 1] \tag{33}$$

NUMERICAL RESULTS

All computations were performed by a CDC-6400 digital computer. The numerical integrations, necessary to obtain the matrix **A** and the expansion coefficients a_n, were done by the method of Romberg (*21*), the accuracy of which was checked by comparing numerical solutions with the analytical integral solutions obtained for limiting cases [Equations (30) to (33)]. The eigenvalues and eigenvectors of the characteristic Equation (25) were determined by the Jacobi method for diagonalization of a real, symmetrix matrix (*22*). The zeros of the first-order Bessel function, available in standard references, were obtained from the text by Watson (*23*). Fifteen-term expansions were used in the solution of the differential equation.

A three-dimensional parametric study was performed in which the following parameters were systematically varied: the rheological parameters, $s = 0.2, 0.4, 0.6, 0.8, 1.0$ for the power law fluid, and $b = 1, 3, 5, 10$ for the Prandtl-Eyring fluid; the reciprocal of the Damköhler Group II, $\kappa = 0, 0.01, 0.05, 0.1, 1.0$; and the Damköhler Group I, $\zeta = 0, 0.02, \cdots 7.0$.

The first six eigenvalues and expansion coefficients for the various cases studied are shown in Tables 1 and 2, respectively.* The values of the first three eigenfunctions evaluated at $\xi = 0.5$ and 1.0 for the cases of $\kappa = 0.01$ and 1.0 are given in Table 3.* The eigenfunctions were normalized by setting $\Xi_n = 1.0$ at $\xi = 0$. Of most practical concern is the variation of the dimensionless bulk concentration with ζ, which is tabulated in Table 4* for the special case of $\kappa = 0.01$; these results are also shown graphically for the power law and Prandtl-Eyring fluids in Figures 1 and 2, respectively. Figure 3 shows the variation of the dimensionless bulk concentration with ζ for a power law fluid of $s = 0.4$ with κ as a parameter.

DISCUSSION OF RESULTS

The eigenvalues, expansion coefficients, and eigenfunctions for the case of a Newtonian fluid for which $s = 1.0$

*Tabular material has been deposited as document No. 01249 with the ASIS National Auxiliary Publications Service, c/o CCM Information Sciences, Inc., 22 W. 34th St., New York 10001 and may be obtained for $2.00 for microfiche or $5.00 for photocopies.

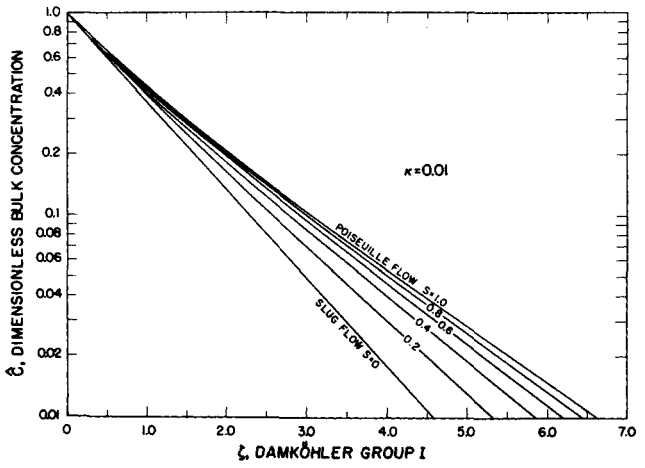

Fig. 1. Dependence of dimensionless bulk concentration upon Damköhler Group I for Ostwald-de Waele fluids at $\kappa = 0.01$.

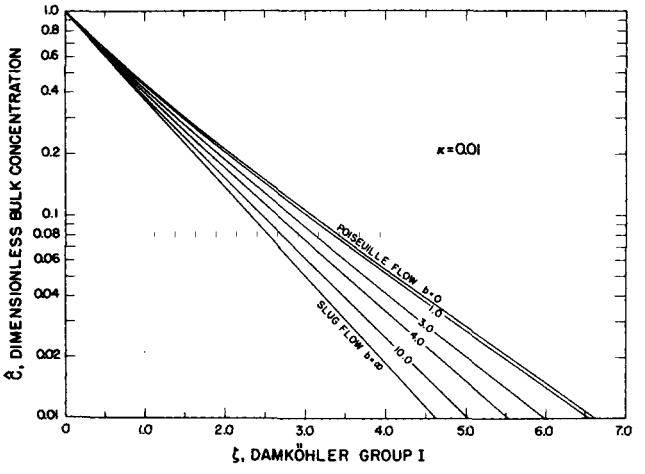

Fig. 2. Variation of dimensionless bulk concentration with Damköhler Group I for Prandtl-Eyring fluids at $\kappa = 0.01$.

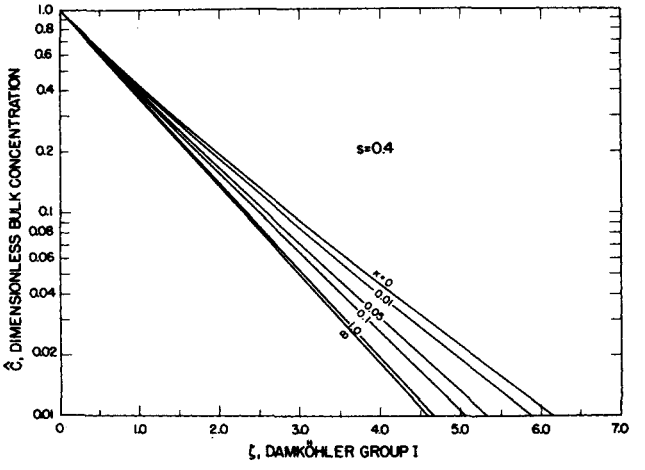

Fig. 3. Effect of Damköhler Group I on dimensionless bulk concentration with κ as a parameter for an Ostwald-de Waele fluid with $s = 0.4$.

compare quite well with the results of previous workers (*7, 8, 10*). Reference to Table 1 shows that the eigenvalues increase monotonically with increasing κ and decreasing psuedoplasticity, that is, increasing s or decreasing b. The results obtained for the dimensionless bulk concentration $\hat{C}$ summarized in Table 4 agree well with literature values for the case of a Newtonian fluid (*1, 7, 8, 10*), thus affording a good test of the accuracy of our calculations.

Several important results may be obtained from Figures 1 and 2. The curves relating the logarithm of $\hat{C}$ with ζ for the various non-Newtonian fluids show a slight deviation from linearity only at small ζ and are all contained within the envelope bounded by Poiseuille flow for which $s = 1.0$ or $b = 0$ and slug flow for which $s = 0$ or $b = \infty$. Indeed, the curves approach complete linearity in the limit of slug flow. While holding ζ constant and as the fluid becomes more pseudoplastic, the bulk concentration of reactant is decreased. Reference to Figure 3 shows that the curves for various values of κ are also very nearly linear and rapidly approach the limiting curve for infinite κ, which is the same limiting curve as is found for slug flow. Further, it may be noted that the bulk concentration is decreased by an increase in κ at fixed ζ. These observations seem reasonable if we consider that increasing the pseudoplasticity of a fluid results in a more blunt velocity profile and a slowing down of the center streamlines, while increasing κ results in an increased rate of diffusion of unreacted species from the center of the reactor to the slower moving fluid near the wall. The overall result of these two effects is to decrease the bulk concentration of reactant or increase the chemical conversion at any axial position in the tube.

It is interesting to examine the behavior of the concentration profile at a given axial position for various non-Newtonian fluids. Figure 4 shows this situation for power law fluids at $\zeta = 5.0$ and $\kappa = 0.01$. It is apparent that the concentration is a maximum at the center, as is to be expected, and decreases with increasing pseudoplasticity. Near the wall, however, the situation is reversed, that is, the concentration increases with increasing pseudoplasticity, the inversion occurring between $6.5 < \xi < 7.8$. For the case of slug flow the concentration profile is constant.

It is necessary to discuss the accuracy of the numerical results that have been presented. Reference to Equation (10) shows that the error incurred by truncating this infinite series at N terms is increased as the dimensionless group $\lambda_n^2\zeta/\beta$ is decreased. From Table 1 it can be seen that the ratio λ_n^2/β is a minimum for the case $\kappa = 0$ and $s = 1.0$, so that the truncation error should increase as we approach $\kappa = 0$, $s = 1.0$ and $\zeta = 0$. A simple estimate of this error was made by comparing the dimensionless bulk concentration calculated with a 15-term expansion of Equation (28) for the case, $\kappa = 0$, $s = 1.0$ and various values of ζ, with the analytic solution obtained by Cleland and Wilhelm (*1*). Error was found in the sixth significant figure at

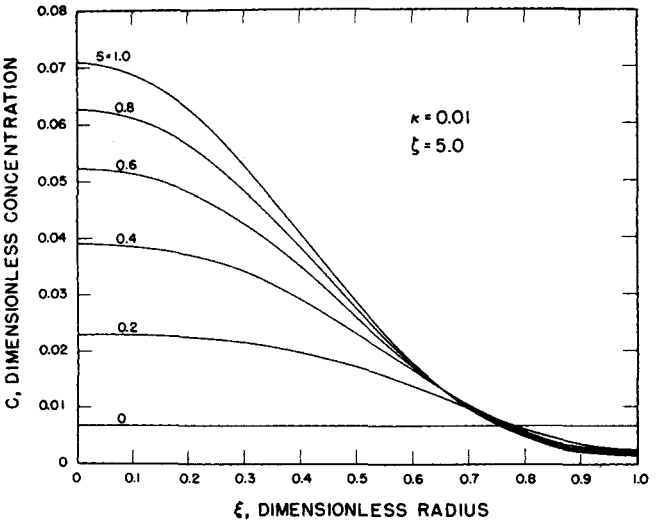

Fig. 4. Concentration profile for $\kappa = 0.01$, $\zeta = 5.0$ for Ostwald-de Waele fluids.

$\zeta = 1.2$, while further downstream at $\zeta = 2.5$ uncertainty was found in only the seventh significant figure. An alternate method to obtain accurate solutions near the entrance region where $\zeta = 0$ would seem to be a similarity solution analogous to that development by Lévêque (*24*) and later extended by Newman (*25*) in the study of the Graetz problem.

Some remarks are in order regarding the suitability of the Galerkin method to the solution of the characteristic Equation (11). A truncation error is incurred by approximating the eigenfunctions by a finite series of N trial functions as in Equation (14). This error is an indication of how well the trial function approximates the eigenfunction Ξ_n. Careful choice of the trial functions as solutions of the auxiliary equation usually simplifies the problem and enhances the accuracy of the results. If such a choice is not possible, other calculational methods such as numerical integration (*8*) may prove to be superior to trial function techniques. Indeed, the best calculational method for a given problem should be determined by a careful comparison of the accuracy and difficulty of implementation of the methods available.

Finally it should also be noted that the axial diffusion term, which has been neglected in Equation (8), becomes important at low Peclet number. Typically, Peclet numbers are large and axial diffusion becomes important only in a small region where $\zeta = 0$ and $\xi = 1.0$. The method of singular perturbations applied by Newman (*26*) in this region for the Graetz problem could be extended to treat the corresponding case here.

SUMMARY

The theoretical treatment of the problem involving an irreversible, first-order chemical reaction in an isothermal, laminar flow tubular reactor was extended to include the flow of non-Newtonian fluids. The characteristic value problem resulting from separation of variables was solved by the method of Galerkin for various Ostwald-de Waele and Prandtl-Eyring fluids. A parametric study was conducted in which the variables s and b, κ, and ζ were systematically varied and the eigenvalues, eigenfunctions, expansion coefficients, bulk concentration, and concentration profile were determined. It was found that near the center of the tube the concentration of reactant decreases with increasing pseudoplasticity, while near the wall the situation is reversed. It was further found that the logarithm of the bulk concentration is very nearly linear with the Damköhler Group I. The effect of increasing the pseudoplasticity of the fluid and increasing the inverse Damköhler Group II is to decrease the bulk concentration of reactant at any axial position.

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NOTATION

a = expansion coefficient
$\mathbf{A}$ = matrix of A_{ij}
b = rheological parameter for Prandtl-Eyring model
$\mathbf{B}$ = matrix of B_{ij}
c = concentration of reactant
$C = c/c_0$ = dimensionless concentration of reactant
$\hat{c}$ = bulk (cup mixing) concentration of reactant
$\hat{C} = \hat{c}/c_0$ = dimensionless bulk concentration of reactant
c_0 = concentration of reactant at $z = 0$
D = molecular diffusivity
$\mathbf{H}$ = matrix of H_{ij}
$\mathbf{I}$ = identity matrix
k = chemical reaction rate constant
N = number of terms in series expansion
p = pressure
r = radial coordinate
R = tube radius
s = rheological parameter for Ostwald-de Waele model
$\mathbf{S}$ = matrix of S_{ij} (transformation matrix)
v = axial velocity
$\langle v \rangle$ = average axial velocity
X_{in} = i^{th} element of the n^{th} eigenvector
$\mathbf{X}$ = eigenvector
$\tilde{\mathbf{X}}$ = transformed eigenvector
z = axial coordinate

Greek Letters

α = zeros of first-order Bessel function
β = constant in generalized velocity expression
γ = trial function
δ_{ij} = Kronecker delta
$\zeta = kz/\langle v \rangle$ = Damköhler Group I
η = apparent viscosity
θ = angular coordinate
$\kappa = D/kR^2$ = (Damköhler Group II)$^{-1}$
λ = eigenvalue
μ, ν, ω = constants in Prandtl-Eyring model
ξ = dimensionless radial coordinate
Ξ = eigenfunction
τ = shear stress
ϕ = dimensionless radial function in generalized velocity expression

Superscript

$( )^T$ = transpose of $( )$

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