Effect of Radial Velocities on Laminar Flow, Tubular Reactor Models

Existing models for laminar flow in tubular reactors neglect radial velocities in the momentum transport equation. The axial velocity distribution is calculated assuming fully developed flow wherein axial velocities are in local equilibrium with the radial viscosity profile. A test of this assumption has been devised using a representative model of the Newtonian viscosity, $\mu(r, z)$, and by reconstructing the radial velocity components using the continuity equation. The assumption of fully developed flow is shown to be a valid approximation for typical polymerization reactions. However, the fact that radial velocities are negligible with respect to momentum transport does not mean they are negligible with respect to convective diffusion of heat and mass. Radial convection is shown to be important for typical polymerization and heat transfer examples.

H. S. McLaughlin,
Ramesh Mallikarjun
and E. B. Nauman
Department of Chemical Engineering
and Environmental Engineering
Rensselaer Polytechnic Institute
Troy, NY 12180

SCOPE

Sound design techniques for laminar flow reactors and heat exchangers must take temperature- and composition-sensitive viscosities into account. A computationally simple technique for doing this emerged in the 1970's (Lynn and Huff, 1971) and has had broad acceptance. In a tubular geometry, the method ignores radial velocities and assumes that axial velocities are in local equilibrium with the radial viscosity profile. In essence, $\mu(r, z)$ is assumed to vary slowly if at all in the z direction. This assumption is not valid in real systems, particularly near the inlet, where viscosities can rapidly change from their initial values. The rapid change in viscosity causes a corresponding change in the velocity profile, which can only be accomplished through radial flow. This paper examines the effect of finite radial velocities and other neglected terms in the axial equation of motion to see if the Lynn and Huff approximation remains valid under physically realistic conditions. It also examines the effects of radial convection on the heat and mass transfer occurring within the reactor. Most authors have ignored radial convection in apparent consistency with the Lynn and Huff approximation. However, the fact that radial velocities are negligible in the equation of motion does not necessarily imply they are negligible in the convective diffusion equation.

CONCLUSIONS AND SIGNIFICANCE

The magnitude of various inertial and viscous terms, neglected under the Lynn and Huff approximation, can be calculated from the continuity equation. These calculations can be made directly from a comprehensive system model or they can be estimated using an exponential viscosity model. The Lynn and Huff approximation is found to be valid for typical reactors and heat exchangers, even for the particularly severe conditions encountered in free radial polymerizations. The main requirement is that the entrance Reynolds number be low, say $Re < 10$.

Radial velocities can be important in the convective diffusion equations even though they are negligible with respect to the axial equation of motion. This is shown through several case studies reflecting physically realistic conditions. It is recommended that radial veloci-

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Correspondence concerning this paper should be addressed to E. B. Nauman.

ties be routinely included in modeling studies of laminar flow reactors and heat exchangers. The computational cost of doing this is fairly small, and it can lead to significant improvements in accuracy.

Introduction

Variable-viscosity, laminar-flow models have become a standard design tool for tubular polymerization reactors. Lynn and Huff (1971) demonstrated the feasibility and utility of numerical simulation. They solved the equations of flow simultaneously with the mass and energy balances in an explicit marching-ahead scheme for a polymerization reaction, generating the velocity, concentration, and thermal profiles for the entire reactor. The tendency of the flow field to elongate and the extreme sensitivity of conversion to the reactor wall temperature were clearly demonstrated.

Lynn and Huff assumed that the flow field would be fully developed throughout the reactor and that radial velocities were negligible. The second assumption is certainly not in conflict with the first, but does not necessarily follow from it. It will be shown that radial velocities are usually negligible with respect to momentum transport in tubular polymerizers but have a significant effect on the radial transfer of heat and mass. The practice of ignoring radial convection has become commonplace in the literature. Lynn and Huff (1971), Wallis et al. (1975), Wyman and Carter (1976), and Castro et al. (1982) all make this assumption. Recently, Hammer (1984) has included radial convection in his model of a vinyl acetate polymerizer, but he retains the assumption of negligible radial velocity in the momentum transport equation.

Equations of Motion

Attention is restricted to the steady, axisymmetric flow of a constant-density, viscous Newtonian fluid. The equation of motion for the axial velocity component is:

$$\rho\left(v_r\frac{\partial v_z}{\partial r}+v_z\frac{\partial v_z}{\partial z}\right) = -\frac{\partial P}{\partial z}+\frac{1}{r}\frac{\partial}{\partial r}\left[r\mu\left(\frac{\partial v_z}{\partial r}+\frac{\partial v_r}{\partial z}\right)\right]+2\frac{\partial}{\partial z}\left(\mu\frac{\partial v_z}{\partial z}\right) \quad (1)$$

This may be written as

$$I_1+I_2=-\frac{\partial P}{\partial z}+V_1+V_2+V_3 \quad (2)$$

where I_1 and I_2 represent inertial terms and V_1, V_2, and V_3 represent viscous terms as defined in Table 1. The Lynn and Huff (1971) approximations effectively assume that I_1, I_2, V_2, and V_3 are all negligible compared to V_1. The equation of motion becomes

$$0=-\frac{dP}{dz}+\frac{1}{r}\frac{d}{dr}\left(r\mu\frac{dv_z}{dr}\right) \quad (3)$$

Table 1. Inertial and Viscous Terms in Axial Equation of Motion

Term	Definition
I_1	$\rho v_r\frac{\partial v_z}{\partial r}$
I_2	$\rho v_z\frac{\partial v_z}{\partial z}$
V_1	$\frac{1}{r}\frac{\partial}{\partial r}\left[r\mu\frac{\partial v_z}{\partial r}\right]$
V_2	$\frac{1}{r}\frac{\partial}{\partial r}\left[r\mu\frac{\partial v_r}{\partial z}\right]$
V_3	$2\frac{\partial}{\partial z}\left[\mu\frac{\partial v_z}{\partial z}\right]$

Using the boundary conditions $v_z = 0$ at $r = R$ and $dv_z/dr = 0$ at $r = 0$ gives

$$\frac{v_z(r)}{\bar{u}}=\hat{v}_z=\frac{R^2\int_r^R\frac{r}{\mu}dr}{2\int_0^R r\int_r^R\frac{r}{\mu}drdr}=\frac{R^2\int_r^R\frac{r}{\mu}dr}{\int_0^R\frac{r^3}{\mu}dr} \quad (4)$$

The axial velocity can thus be found from knowledge of $\mu(r, z)$. The above formulation allows μ to be a rapidly varying function of r, but it must vary slowly if at all in the z direction. Otherwise, v_r and derivatives such as $\partial v_z/\partial z$, which are ignored in Eq. 3, will become important.

Whenever μ changes in the axial direction, some radial velocities must exist. Even though they may be negligible with respect to Eq. 3, they can be calculated from the continuity equation:

$$\frac{1}{r}\frac{\partial}{\partial r}(rv_r)+\frac{\partial v_z}{\partial z}=0 \quad (5)$$

which integrates to

$$v_r=-\frac{1}{r}\int_0^r r\left(\frac{\partial v_z}{\partial z}\right)dr \quad (6)$$

Thus the assumption of fully developed flow does not preclude calculation of v_r and its subsequent inclusion in the convective diffusion equations. Calculation of v_r also allows evaluation of the neglected terms in Eq. 2, i.e., I_1, I_2, V_2, and V_3. If these terms are always one or more order of magnitude smaller than V_1, the assumption of fully developed flow is justified.

Ordinarily, $\mu(r, z)$ is obtained from a physical properties correlation involving temperature and composition. In essence, Eq. 4 and 6 must be solved simultaneously with the convective diffusion equations for heat and mass. This was done for the specific polymerization and heat transfer examples to be discussed in a subsequent section. However, a different approach is useful to test the validity of Eq. 4. We assume viscosity varies within the reactor as

$$\mu = \mu_o \exp(\alpha r z) = \mu_o \exp(R^2 \alpha \mathbf{r} \mathbf{z}) \qquad (7)$$

where $\mathbf{r} = r/R$ and $\mathbf{z} = z/R$. This functional form is broadly representative of the viscosity profile in tubular polymerizers. When the fluid begins reacting at $\mathbf{z} = 0$, it has a uniform viscosity and a parabolic velocity profile. Polymerization is highest at the wall, and this gives rise to more viscous fluid near the wall. The axial velocity decreases at the wall and must accelerate at the centerline to maintain the same total flow down the tube. The lowered velocities near the wall lead to still further polymerization and larger viscosities. This process builds on itself, giving highly elongated velocity profiles that can eventually become hydrodynamically unstable. Figures 1 and 2 show axial and radial velocity profiles for the case $R^2\alpha = 1$ and $1 \leq \mathbf{z} \leq 25$. Note that we have assumed an exponential dependence of viscosity on radial position. We assert that this is broadly representative of bulk polymerizations in tubular reactors. It allows analytical solutions for v_z and v_r, yet gives elongated velocity profiles typical of those calculated using detailed models of bulk polymerizations. By varying α and $\mathbf{z}$, different extents and rates of elongation can be studied. In essence, this corresponds to varying the polymerization rate. Note also that the usual entrance length problem, such as a transition from a flat to a parabolic profile, has been ignored. In single-tube polymerizers, the position $\mathbf{z} = 0$ typically corresponds to the beginning of a heated region rather than to the hydrodynamic entry point. In multitubular reactors, a flat to parabolic transition may be plausible, but the entrance length will be only about one tube radius for the low Reynolds number flows being considered here.

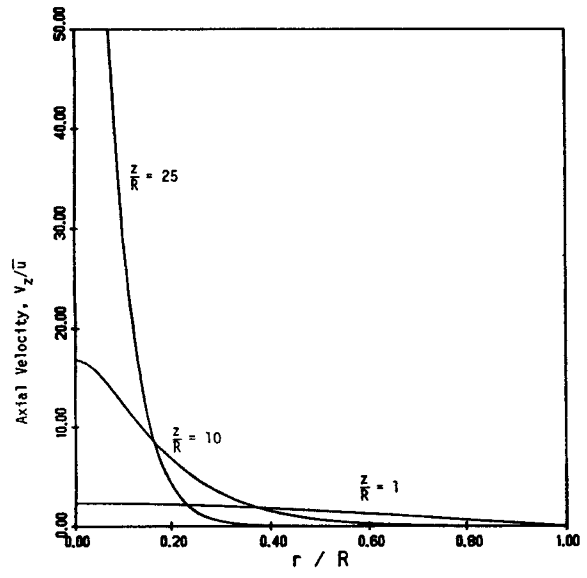

Figure 1. Axial velocity profiles for exponential viscosity model, $R^2\alpha = 1.0$.

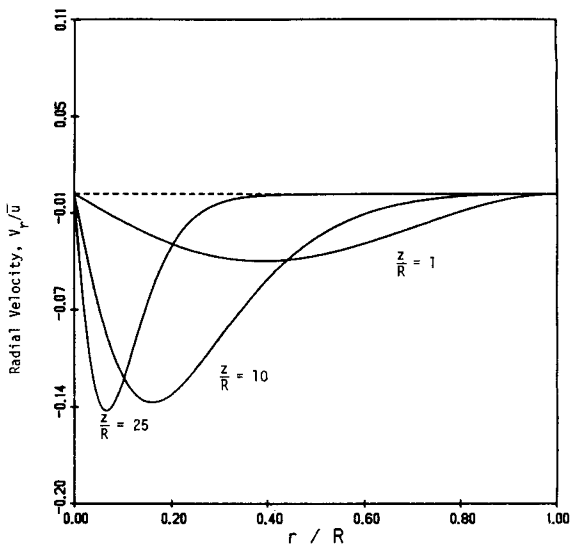

Figure 2. Radial velocity profiles for exponential viscosity model, $R^2\alpha = 1.0$.

Table 2 gives definitions for the ratios to V_1 of the various inertial and viscous terms. For the exponential viscosity model of Eq. 7, the magnitudes of these ratios depend only on the viscosity parameter, α, and the inlet Reynolds number, Re. The scaling factors shown in Table 2 were derived from the analytical solution of Eqs. 3 through 7. They allow a single set of master curves to be used for all possible combinations of α and $\mathbf{z}$. Figure 3 gives master curves with $(Re)_m = 1$, $R^2\alpha_m = 1$, and $1 < \mathbf{z}_m < 25$. The curves for $\mathbf{z}_m < 1$ are very similar to those for $\mathbf{z}_m = 1$, and no serious error is introduced by using the $\mathbf{z}_m = 1$ curve for $\mathbf{z}_m < 1$. To find an inertial or viscous ratio corresponding to some $\mathbf{z}$, Re, and $R^2\alpha \neq 1$, enter the master curves at $\mathbf{z}_m = \mathbf{z}\alpha/\alpha_m$. This determines a ratio, unscaled for α, at the same dimensionless vis-

Table 2. Ratios and Scaling Factors for Inertial and Viscous Terms

Ratio	Definition in Dimensionless Form	Scaling Factor for Exponential Viscosity Model
$\frac{\lvert I_1\rvert}{\lvert V_1\rvert}$	$\frac{Re}{2}\frac{\left\lvert r\hat{v}_r \frac{\partial \hat{v}_z}{\partial r}\right\rvert}{\left\lvert \frac{\partial}{\partial r}\left(r\hat{\mu}\frac{\partial \hat{v}_z}{\partial r}\right)\right\rvert}$	$\frac{Re\,\alpha}{(Re)_m\,\alpha_m}$
$\frac{\lvert I_2\rvert}{\lvert V_1\rvert}$	$\frac{Re}{2}\frac{\left\lvert r\hat{v}_z \frac{\partial \hat{v}_z}{\partial z}\right\rvert}{\left\lvert \frac{\partial}{\partial r}\left(r\hat{\mu}\frac{\partial \hat{v}_r}{\partial r}\right)\right\rvert}$	$\frac{Re\,\alpha}{(Re)_m\,\alpha_m}$
$\frac{\lvert V_2\rvert}{\lvert V_1\rvert}$	$\frac{\left\lvert \frac{\partial}{\partial r}\left(r\hat{\mu}\frac{\partial \hat{v}_z}{\partial r}\right)\right\rvert}{\left\lvert \frac{\partial}{\partial r}\left(r\hat{\mu}\frac{\partial \hat{v}_z}{\partial r}\right)\right\rvert}$	$(\alpha/\alpha_m)^2$
$\frac{\lvert V_3\rvert}{\lvert V_1\rvert}$	$\frac{\left\lvert 2\frac{\partial}{\partial z}\left(\mu\frac{\partial \hat{v}_z}{\partial z}\right)\right\rvert}{\left\lvert \frac{\partial}{\partial r}\left(r\hat{\mu}\frac{\partial \hat{v}_z}{\partial r}\right)\right\rvert}$	$(\alpha/\alpha_m)^2$

$Re = 2\rho\bar{u}R/\mu_0$; $\hat{v}_z = v_z/\bar{u}$; $\hat{v}_r = v_r/\bar{u}$; $\hat{\mu} = \mu/\mu_0$

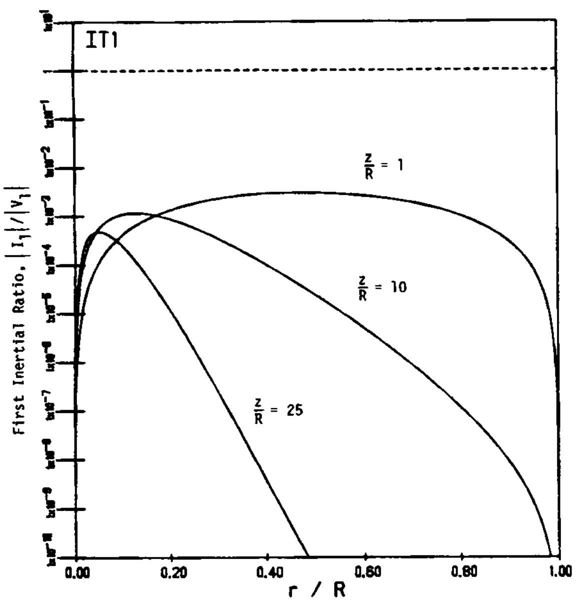

Figure 3a. Master curves of first inertial ratio for exponential viscosity model, $R^2\alpha_m = 1.0$.

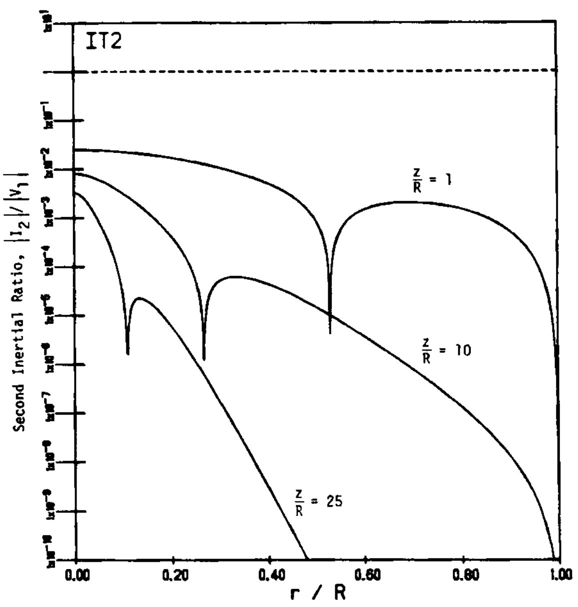

Figure 3b. Master curves of second inertial ratio for exponential viscosity model, $R^2\alpha_m = 1.0$.

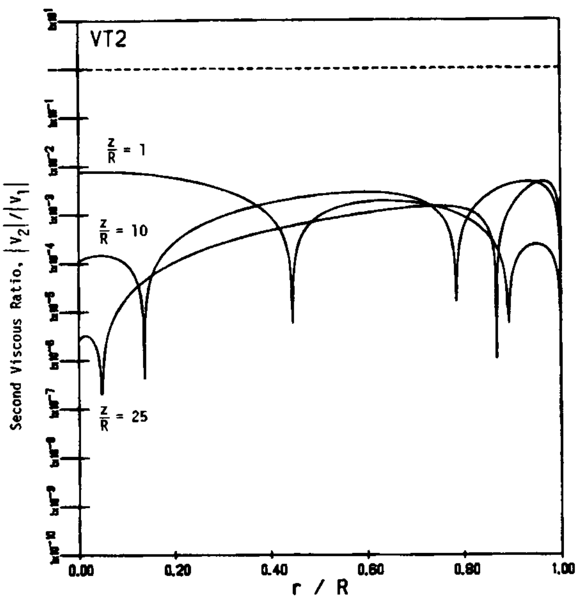

Figure 3c. Master curves of second viscous ratio for exponential viscosity model, $R^2\alpha_m = 1.0$.

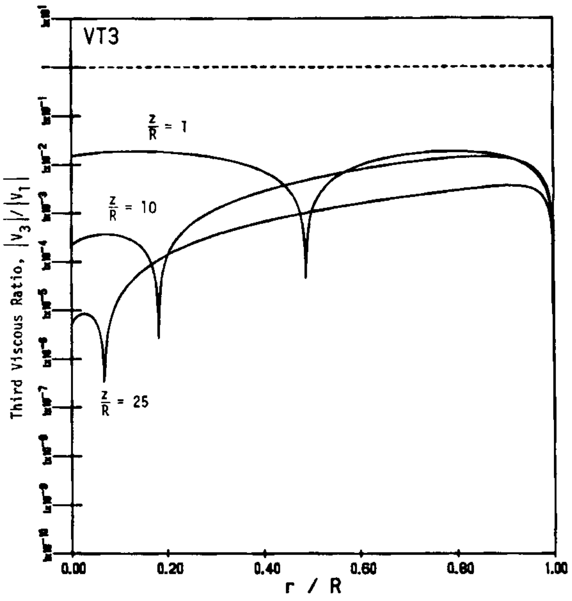

Figure 3d. Master curves of third viscous ratio for exponential viscosity model, $R^2\alpha_m = 1.0$.

cosity profile. The unscaled ratio is then multiplied by the scaling factor to obtain the inertial or viscous ratios at the specified values of Re, $R^2\alpha$, and z. Note that absolute values are plotted in Figure 3. The apparent discontinuities in slope shown in Figures 3b, c, d are due to sign changes in one of the underlying variables. The individual variables are all continuous, with continuous first derivatives.

Suppose it is desired to test the assumption of fully developed flow in a tubular polymerizer with $Re = 2$ and a relatively fast reaction such that $R^2\alpha = 20$. We begin with the first inertial ratio, $|I_1|/|V_1|$. The size of this ratio will vary with both z and r. Pick $z = 0.5$ as a point for evaluation. Figure 3a is used with $z_m = z\alpha/\alpha_m = 0.5(20)/1 = 10$. For $z_m = 10$, Figure 3a shows a maximum value for $|I_1|/|V_1|$ of 0.001 at $r = 0.02$. This is scaled by a factor of $Re\alpha/(Re_m\alpha_m) = 40$ to give $|I_1|/|V_1| = 0.04$. Similarly, the maximum values for $|I_2|/|V_1|$, $|V_2|/|V_1|$, and $|V_3|/|V_1|$ are 0.36, 0.6, and 1.1, respectively. Thus the case of $Re = 2$, $R^2\alpha = 20$, and $z = 0.5$ violates the assumption of fully developed flow.

The maxima for the various inertial and viscous ratios occur at small values of z for which the curves for $z = 1$ are a reasonable approximation. The values of the maximum ratios depend on α and Re: $|I_1|/|V_1| < 0.1$ if $\alpha Re < 16$; $|I_2|/|V_1| < 0.1$ if $\alpha Re < 2.5$; $|V_2|/|V_1| < 0.1$ if $\alpha < 3$; $|V_3|/|V_1| < 0.1$ if $\alpha < 1.8$. Thus the conditions $\alpha Re < 2.5$ and $\alpha < 1.8$ ensure that the neglected terms are all an order of magnitude smaller than V_1.

These conditions are satisfied in tubular polymerizers which typically have entrance Reynolds numbers less than 10. This allows $\alpha = 0.25$ and $\mu_{wall}/\mu_o = e^{12.5} = 2.7 \times 10^5$ at a distance of only 25 diameters from the inlet.

Convective Diffusion Equations

Convective diffusion of some component A is modeled by

$$v_z \frac{\partial a}{\partial z} + v_r \frac{\partial a}{\partial r} = D_A \left[\frac{1}{r} \frac{\partial a}{\partial r} + \frac{\partial^2 a}{\partial r^2} \right] + R_A \tag{8}$$

and convective diffusion of heat is given by

$$v_z \frac{\partial T}{\partial z} + v_r \frac{\partial T}{\partial r} = \alpha_T \left[\frac{1}{r} \frac{\partial a}{\partial r} + \frac{\partial^2 a}{\partial r^2} \right] + \frac{\Delta H R_A}{\rho c_v} \tag{9}$$

Equations of this sort have been employed in all the previously cited polymerization models. At issue is whether the v_r term should be retained. Most studies have dropped it, but in what follows it will be shown that neglecting v_r leads to serious errors in the temperature, composition, and velocity fields.

Radial velocities are negligible with respect to the equation of motion in the following examples, i.e., I_1, I_2, V_2, and V_3 are small compared to V_1. Thus v_z may be calculated from Eq. 4. Radial velocities were reconstructed from Eq. 6. Temperatures and compositions were calculated using Eqs. 8 and 9 both retaining and neglecting the v_r terms. Viscosities were calculated from an appropriate correlation. Note that including v_r in the convective diffusion equations affects μ and thus indirectly v_z even though v_r has no direct affect on v_z.

Example 1. isothermal polymerizer with second-order kinetics

Biesenberger (1965) showed that stepwise condensation polymerizations with equal reactivity can be modeled as a second-order decay of the fraction unreacted of functional groups, ϕ. For this study, the reactor was regarded as isothermal, and the viscosity was treated as a function of the conversion only. The viscosity increases 100-fold with total conversion in the following nonlinear fashion:

$$\mu = 1 + 100\,(1 - \phi)^3 \tag{10}$$

In the experience of the authors, this conversion-viscosity behavior is representative of condensation polymerizations in solvents. Molecular diffusion was assumed to be negligible so that Eq. 8 reduced to

$$v_z \frac{\partial \phi}{\partial z} + v_r \frac{\partial \phi}{\partial r} = -k\phi^2 \tag{11}$$

Table 3 gives the results of the simulation. There are significant differences in overall conversion due to radial convection. Table 4 confirms that the various inertial and viscous ratios are all small. These ratios were calculated numerically and correspond to $Re = 1$ and $L/R = 100$. The convective diffusion equation was solved using a fully explicit finite difference in this simple example and the one that follows. The method of lines was employed for the detailed simulation in example 3.

Table 3. Equal-Reactivity Polycondensation with Variable Viscosity

	Fraction Unreacted, ϕ		Centerline Velocity $\hat{v}_z = v_z/\bar{u}$	
$k\bar{t}$	$v_r = 0$	$v_r \neq 0$	$v_r = 0$	$v_r \neq 0$
0	1.000	1.000	2.00	2.00
1	0.652	0.672	6.73	5.93
2	0.443	0.508	5.00	4.61
3	0.329	0.398	3.67	3.63
4	0.262	0.322	3.05	3.09
5	0.218	0.268	2.74	2.79

Example 2. laminar heat exchanger with first-order reaction

This example models a constant wall temperature heat exchanger in which a first-order reaction is occurring. Viscosity is a function of temperature alone and is given by a generalized correlation (Perry and Chilton, 1975) for liquids

$$\mu = \exp\,(17.4 - 0.20\,T + 0.00072\,T^2) \tag{12}$$

where μ is in centipoise (mPa·s) and T in °C. The simulation involved cooling a hot, reactive liquid with a 50°C difference between the entering liquid and the cool walls. The viscosity changes by a factor of 100 in this range. A temperature-insensitive, first-order reaction occurred with $k\bar{t} = 0.3$. Radial diffusion of heat was governed by $\alpha_T\bar{t}/R^2 = 0.4$. Results are given in Table 5. Conversions calculated with and without radial convection are again quite different.

Example 3. a styrene polymerization

This example used a complex model of styrene polymerization in a 5 m long, 0.0127 m diam. tubular reactor having a wall temperature of 120°C, an inlet temperature of 130°C, and a mean residence time of 1 h. The model is generally similar to those used in previously cited studies of styrene polymerization. Details of the model are given elsewhere (Mallikarjun, 1985). Figure 4 shows exit composition distributions. Again, radial convection plays an important role in reactor performance even though it has a negligible effect on the axial equation of motion.

Conclusions

The assumption of fully developed axial flow is reasonable for bulk polymerization reactors. Quite large radial gradients in viscosity can develop and these lead to substantially elongated velocity profiles. However, the resulting radial velocities and axial accelerations have only a small effect on the axial equation of motion. The changes in radial viscosities and axial velocities occur slowly enough (i.e., at large enough values of z) and at low

Table 4. Inertial and Viscous Terms for Polycondensation Example

$z = z/R$	$\lvert I_1 \rvert / \lvert V_1 \rvert$	$\lvert I_2 \rvert / \lvert V_1 \rvert$	$\lvert V_2 \rvert / \lvert V_1 \rvert$	$\lvert V_3 \rvert / \lvert V_1 \rvert$
2.5	6.6×10^{-3}	3.0×10^{-2}	4.9×10^{-3}	1.5×10^{-2}
10	2.9×10^{-4}	2.4×10^{-3}	2.5×10^{-3}	7.6×10^{-4}
25	2.0×10^{-4}	7.9×10^{-4}	1.1×10^{-4}	6.8×10^{-5}

Table 5. First-Order Reaction within a Cooling Heat Exchanger

Axial Position z/L	Centerline Velocity $\hat{v}_z = v_z/\bar{u}$		Mixing Cup Temp. °C		Fraction Unreacted	
	$v_r = 0$	$v_r \neq 0$	$v_r = 0$	$v_r \neq 0$	$v_r = 0$	$v_r \neq 0$
0	2.00	2.00	100.0	100.0	1.000	1.000
0.2	5.36	5.30	88.5	88.1	0.948	0.951
0.4	5.30	5.20	80.5	80.9	0.896	0.911
0.6	4.72	4.73	73.9	75.2	0.846	0.874
0.8	4.05	4.19	68.5	70.4	0.795	0.839
1.0	3.50	3.70	64.3	66.3	0.748	0.805

enough Reynolds numbers that the inertial and secondary viscous terms are several orders of magnitude smaller than the primary viscous term, $(1/r)\partial(r\mu dv_z/\partial r)/\partial r$, which corresponds to fully developed flow.

The exponential viscosity model as used here provides a simple means for estimating the magnitude of the various inertial and viscous terms in the axial equation of motion. The results of a computer simulation can be used to find aproximate values for $R^2\alpha$ and z. This estimation can be based either on the calculated viscosity profile using Eq. 7, or on the extent of velocity elongation using Figure 1. The master curves in Figure 3 can then be used to estimate magnitudes for all the inertial and viscous terms that were neglected under the assumption of fully developed flow. This approach is simpler and is computationally expedient compared to the alternative of calculating the neglected terms using the continuity equation.

Although usually negligible in the axial equation of motion, the radial velocity component can have a significant effect on the convective diffusion of heat and mass. Radial convection will occur whenever the velocity profile is distorted, but reaction conditions that lead to an initial elongation of the velocity profile typically have more effect than conditions that lead to initial flattening. The examples in the study illustrate situations of polymerization and cooling at the wall where the profile initially elongates but returns asymptotically to the parabolic distribution. Errors made by neglecting radial convection while the profile is elongating will be partially compensated by errors made while the profile is returning to parabolic. There remains, however, a substantial, cumulative error in the extent of reaction.

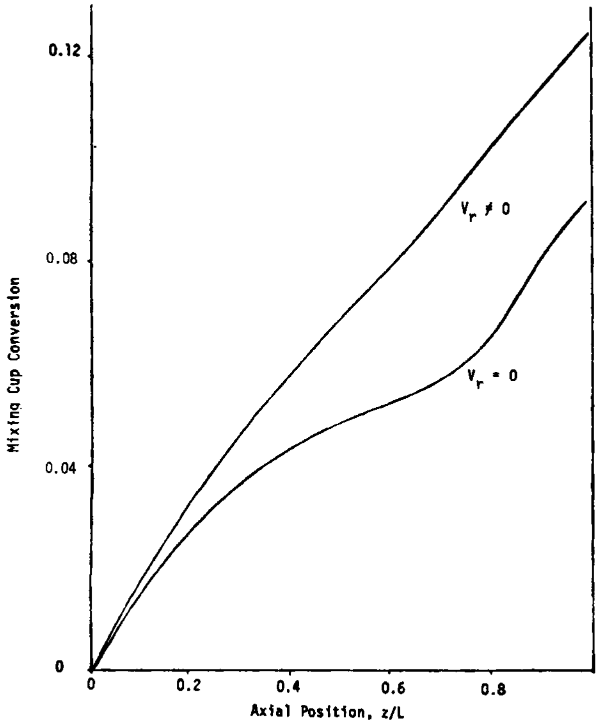

Figure 4. Effect of radial convection on conversion for a styrene polymerization.

Radial convection does occur in practical reactor designs, and its effects can be significant. The radial velocity components are relatively easy to calculate from Eq. 6 and can be readily incorporated into Eqs. 8 and 9 when these are solved by the usual numerical schemes. Thus radial convection should be included routinely in the modeling of laminar tubular reactors and heat exchangers.

Notation

a = concentration of a reactive component
c_v = heat capacity
D = diffusivity
I_1, I_2 = inertial terms, see Table 1
k = reaction rate constant
L = length of reactor
m = subscript denoting value from master curve
P = pressure
r = radial coordinate
$\mathbf{r}$ = dimensionless radial coordinate, r/R
R = radius of tube
$\mathbf{R}$ = reaction rate
Re = Reynolds number at inlet of reactor
$\bar{t}$ = mean residence time
T = temperature
$\bar{u}$ = mean axial velocity
v_r = radial velocity component
$\hat{v}_r$ = normalized radial velocity, $v_r/\bar{u}$
v_z = axial velocity component
$\hat{v}_z$ = normalized axial velocity, $v_z/\bar{u}$
V_1, V_2, V_3 = viscous terms, see Table 1
z = axial coordinate
$\mathbf{z}$ = dimensionless axial coordinate, z/R

Greek letters

α = constant in exponential viscosity model
α_T = thermal diffusivity
ΔH = heat of reaction
μ = viscosity
μ_o = viscosity at tube inlet
ϕ = unreacted fraction of end groups
ρ = density

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