Influence of Secondary Flow on Diffusion with Reaction

Coiling of a tube produces a profound change in the internal flow patterns due to the generation of a centrifugally driven secondary flow. This change strongly influences the transport processes in a coiled tube. The present paper investigates the influence of such secondary flow on a process of convective diffusion with reaction. The findings have an important bearing on a better understanding of a coiled tube reactor.

R. A. MASHELKAR

Chemical Engineering Division
National Chemical Laboratory
Pune 411008, India

C. V. VENKATASUBRAMANIAN

Larsen and Toubro Limited
Powai Works, Bombay 400072, India

SCOPE

A fluid traveling in a coiled tube is subjected to a centrifugal force, which gives rise to a secondary flow. This flow promotes orderly mixing and augments transfer rates. A coiled tube therefore serves as an efficient device for heat transfer, as a membrane oxygenator, as a reverse osmosis device, and as a gas-liquid contactor. It can be used as an efficient chemical reactor too, since the secondary flow narrows down the residence time distribution or the extent of axial dispersion.

Although coiled tubes have been used rather commonly as chemical reactors in industry, there have been no prior studies where the complete problem of convective diffusion with reaction in coiled tubes has been examined. A recent study by Southwick and Seader (1981) provides an elegant experimental demonstration of the advantages of secondary flow in enhancing the rates of reaction. Other experimental studies have investigated axial dispersion in coiled tubes (Janssen, 1976; Nauman, 1977; Trivedi and Vasudeva, 1975; Ranade and Ulbrecht, 1981). Such studies are helpful in describing the performance of a coiled tube chemical reactor in terms of a lumped parameter axial dispersion model, but this does not provide information on the detailed spatial concentration distribution.

It is clear that there are no prior fundamental studies of the problem of convective diffusion with reaction in a flow field that is dominated by a secondary flow. The object of the present work was therefore to analyze this problem so as to gain a depth of knowledge of the influence of secondary flow on concentration distribution as well as the bulk average concentration in a reacting system. Both Newtonian and non-Newtonian fluids are considered, the latter being an important problem since the process of thermal pasteurization of non-Newtonian liquid foods is usually modeled as a process of convective diffusion with reaction. In particular, we have tried to elucidate the role of relative interaction between the shear thinning property of the fluid, the operational parameters governing the secondary flow, and the reaction parameter itself.

CONCLUSIONS AND SIGNIFICANCE

A numerical solution of the problem of convective diffusion with reaction in a coiled tube under low Dean number conditions is presented. The influence of operational variables which govern the intensity of secondary flow (such as Reynolds number and curvature ratio) and of the reaction parameter α (which provide a relative measure of the chemical reaction and molecular diffusion) has been systematically examined. It is shown that the presence of even a mild secondary flow can have a dramatic influence on the local concentration distribution. Thus, for instance, for some specific parameter values ($\alpha = 10$, $De = 10$, $Re = 100$, $Sc = 1{,}000$, $\xi = 0.015$) the ratio of maximum to minimum local concentration in a straight tube reactor is 12.4, whereas that in the case of a curved tube reactor is barely 2.8 (horizontal plane) and 3.82 (vertical plane). Secondary flow reduces the effective length of the reactor, producing only minor additional pressure drop. Again, for some specific parameter values ($\alpha = 100$, $De = 10$, $Re = 100$, $Sc = 1{,}000$), it is shown that for reaching 99% conversion, a straight tube reactor is 32% longer than a coiled tube reactor. Considering the additional length of the straight tube reactor, the coiled tube reactor actually operates with 24% less pressure drop.

Because the results are likely to be useful for thermal pasteurization of non-Newtonian liquid foods, the influence of pseudoplasticity has also been examined. The effective flattening provided by pseudoplasticity is shown to dominate over the cross-sectional mixing provided by the secondary flow.

Correspondence concerning this paper should be directed to R. A. Mashelkar.

BACKGROUND

A fluid element traveling in a curved tube is subjected to a centrifugal force resulting in a secondary flow perpendicular to the direction of the main axial flow (Figure 1). Such secondary flows promote orderly mixing and augment transport rates. Therefore, there has been a considerable interest among engineers to exploit coiled tube as an efficient mass or heat transfer contactor. An early review by Srinivasan et al. (1967) summarized the significant activity that took place in this area up to the mid-1960s. Several investigations have appeared over the last decade which have examined both theoretically and experimentally the use of a coiled tube in a variety of situations. These include heat transfer (Janssen and Hoogendoorn, 1978), oxygenation using membrane devices (Weissman and Mockros, 1968), reverse osmosis (Nunge and Adams, 1973), gas-liquid contacting (Kasturi and Stepanek, 1972), and others. In a majority of these cases there have been observations of significant improvement in transport rates due to coiling.

A coiled tube can be used as an efficient chemical reactor. Recent experimental studies by Seader and Southwick (1981) demonstrate rather clearly that such advantages arise due to the fact that the prevailing secondary flow promotes cross-sectional mixing and therefore narrows down the residence time distribution (RTD) or the extent of axial dispersion.

The majority of studies appearing in the literature examine aspects of RTD and axial dispersion in coiled tubes (Ruthven, 1971; Trivedi and Vasudeva, 1975; Janssen, 1976; Nauman, 1977). However, the use of RTD or dispersion models has limitations in terms of the range of their applicability. Additionally, such lumped parameter models predict the averaged-out concentrations at the exit of a reactor, not the details of the concentration distribution within the reactor.

There is no prior study in the literature which examines the influence of secondary flow on the details of the concentration field. We therefore decided to examine the problem of convective diffusion with reaction in a coiled tube through which a Newtonian or a non-Newtonian fluid may flow under the so called low Dean number condition.

Prior Studies on Diffusion with Reaction in Non-Newtonian Fluids

The problem of convective diffusion with reaction in a Newtonian fluid flowing through a tube has been well studied in the literature. The importance of studies in diffusion with or without reaction in non-Newtonian fluids has been reviewed by Astarita and Mashelkar (1977). The incentive to examine the problem of diffusion with first-order reaction in non-Newtonian fluids is seen in the context of thermal pasteurization of non-Newtonian liquid foods. Such processes can be treated as normal kinetic processes. Indeed the death rate of microorganisms is proportional to the population density of the microorganisms and therefore linear kinetics can be expected to hold. This problem also has a bearing on tubular polymerization reactors. However, phenomena such as the enormous viscosity rise in a reacting polymerization mass followed by a local rise in temperature due to high exothermicity dominates the performance of a tubular polymerization reactor (Lynn and Huff, 1971).

Note that the theoretical investigations on a first-order reaction occurring in non-Newtonian fluids flowing through a straight tube have been undertaken previously (Homsy and Strohman, 1971; Mashelkar, 1973; Mohan et al., 1975), but the corresponding problem in the presence of secondary flow has not been investigated at all.

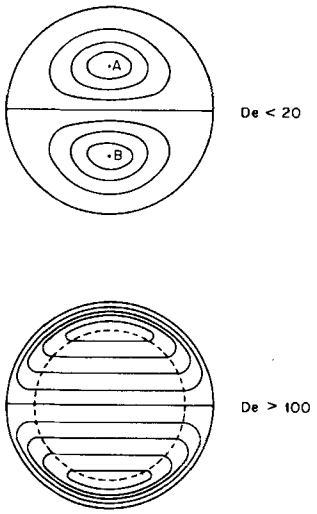

Figure 1. Streamlines of secondary flow field in a coiled tube at low Dean numbers, *De* < 20, and at high Dean numbers, *De* > 100.

HYDRODYNAMICS IN COILED TUBES

To obtain a solution of the problem of convective diffusion with reaction in a coiled tube, the details of the hydrodynamics need to be known. A brief review concerning the prior studies on hydrodynamics in coiled tubes is in order.

Dean (1927, 1928) was the first to investigate theoretically the secondary flow in coils. Many fluid mechanical analyses which make use of analytical, semi-analytical, and numerical techniques (e.g., McConoloque and Srivastava, 1968; Greenspan, 1973; Joseph et al., 1975) have appeared in the literature so far as flow of Newtonian fluids is concerned. The problem of hydrodynamics in non-Newtonian fluids is more complex. The previous studies of hydrodynamics in coiled tubes include investigations of purely viscous non-Newtonian fluids flowing under low Dean number conditions (Raju and Rathna, 1970), viscoelastic fluids flowing under low Dean number conditions (Barnes and Walters, 1969), purely viscous fluids and viscoelastics fluids flowing under high Dean number conditions (Mashelkar and Devarajan, 1976a, 1976b), and purely viscous fluids flowing under turbulent conditions (Mashelkar and Devarajan, 1977). In the present work, we will be concerned with the velocity profiles presented by Raju and Rathna (1970) for the flow of a power-law fluid in a coiled tube under the conditions of low Dean numbers.

Governing Equations

The system of coordinates employed is shown in Figure 2; a is the radius of the tube, and R is the radius of curvature. The flow occurs in the increasing ϵ direction. The velocity vector V has components U, V and W in the r, ϕ and ϵ directions, respectively.

We shall further assume that the influence of axial convection is much stronger in comparison to that of axial molecular diffusion. The contribution of axial diffusion terms could be neglected for $Pe(= aU/D) > 1{,}000$ (Ananthkrishnan et al., 1965), a condition easily satisfied with liquids. We also assume concentration independent diffusivity (D).

Under the above assumptions the convective diffusion equation in the cylindrical coordinate system can be expressed as:

$$\frac{1}{r}\cdot\frac{\partial(rUc)}{\partial r}+\frac{1}{r}\cdot\frac{\partial(Vc)}{\partial\phi}+\frac{\partial(Wc)}{\partial Z^*} = D\left(\frac{\partial^2 c}{\partial r^2}+\frac{1}{r}\frac{\partial c}{\partial r}+\frac{1}{r^2}\cdot\frac{\partial^2 c}{\partial\phi^2}\right)-kc \quad (1)$$

Equation 1 is made nondimension by using

$$\theta = C/C_I,\ \rho=\frac{r}{a},\ Z=\frac{Z^*}{\left(\frac{a^2U_{avg}}{\nu}\right)},\ u=\frac{aU}{\nu},\ v=\frac{aV}{\nu}$$

$$w=\frac{W}{U_{avg}}$$

Then Eq. 1 reduces to

$$\frac{1}{\rho}\cdot\frac{\partial(u\theta)}{\partial\rho}+\frac{1}{\rho}\cdot\frac{\partial(v\theta)}{\partial\phi}+\frac{\partial(w\theta)}{\partial Z}=\frac{1}{Sc}\left(\frac{\partial^2\theta}{\partial\rho^2}+\frac{1}{\rho}\cdot\frac{\partial\theta}{\partial\rho}+\frac{1}{\rho^2}\cdot\frac{\partial^2\theta}{\partial\phi^2}\right)-\frac{\alpha}{Sc}\theta \quad (2)$$

α is a reaction parameter defined as $\alpha = ka^2/D$ and Sc is the Schmidt number ($Sc = \nu/D$). Assume that the fluid under consideration is an Ostwald-de-Waele power-law fluid. Then

$$\tau = -m(\Delta{:}\Delta)^{n-1/2}\,\Delta \quad (3)$$

Raju and Rathna (1970) have solved the problem of flow of a power law fluid in a coiled tube for the case of low Dean numbers. The individual velocity components are given as

$$\frac{U}{U_{avg}} = f_1(n)\,Re\left(\frac{a}{R}\right)\sin\phi[A(n)+B(n)\rho^{s-1}+C(n)\rho^{(2n+2/n)}-D(n)\rho^{(3n+3/n)}]$$

$$\frac{V}{U_{avg}} = f_2(n)\,Re\left(\frac{a}{R}\right)\cos\phi\left[A(n)+sB(n)\rho^{s-1}+\left(\frac{3n+2}{n}\right)C(n)\rho^{(2n+2/n)}-\left(\frac{4n+3}{n}\right)D(n)\rho^{(3n+3/n)}\right] \quad (5)$$

$$\frac{W}{U_{avg}} = \left(\frac{3n+1}{n+1}\right)(1-\rho^{n+1/n})+\left(\frac{a}{R}\right)w_1 \quad (6)$$

where

$$w_1 = \sin\phi\left[f_2(n)\,Re^2\rho^{1/n}\left\{30A(n)(1-\rho^{n+1/n})+\frac{60B(n)(n+1)^2}{(ns+1)(ns+2+n)}(1-\rho^{(s+1/n)})+5C(n)(1-\rho^{(3n+3/n)})-3D(n)(1-\rho^{(4n+4/n)})\right\}-(1-\rho)+\left(\frac{3n^2+7n+4}{2n(3n+1)}\right)(1-\rho^{1/n})+\left(\frac{3n^2-5n-4}{2n(3n+1)}\right)(1-\rho^{(2n+1/n)})\right] \quad (7)$$

$$s = \frac{n+1}{2n}+\frac{\sqrt{(\sqrt{17}n-1)^2+2n(\sqrt{17}-1)}}{2n} \quad (8)$$

$$A(n) = n^3\frac{[ns(21n^3+53n^2+38n+8)-(60n^4+185n^3+200n^2+92n+15)]}{12(n+1)(1-s)(2n+1)(3n+1)(4n^2+9n+3)(n^2+4n+1)} \quad (9)$$

$$B(n) = \frac{n^3(13n^3+31n^2+23n+5)}{4(1-s)(2n+1)(3n+1)(n^2+4n+1)(4n^2+9n+3)} \quad (10)$$

$$C(n) = \frac{n^4}{4(n+1)(3n+1)(n^2+4n+1)} \quad (11)$$

$$D(n) = \frac{n^4}{12(n+1)(2n+1)(4n^2+9n+3)} \quad (12)$$

$$Re = \frac{(2a)^n U_{avg}{}^{2-n}\rho_L}{m} \quad (13)$$

$$f_1(n) = \left(\frac{1}{n+1}\right)\left(\frac{3n+1}{4n}\right)^{1-n}2^{\left(\frac{3+11n-6n^2}{2n}\right)} \quad (14)$$

$$f_2(n) = \frac{1}{120n\left[2^{2(3n^2-4n-1)/n}\left(\frac{3n+1}{4n}\right)^{2n}(n+1)^2\right]} \quad (15)$$

At $r = a$, $\phi = -\pi/2$ represents the inner wall and $\phi = +\pi/2$ represents the outer wall.

Now symmetry consideration leads to a B.C.

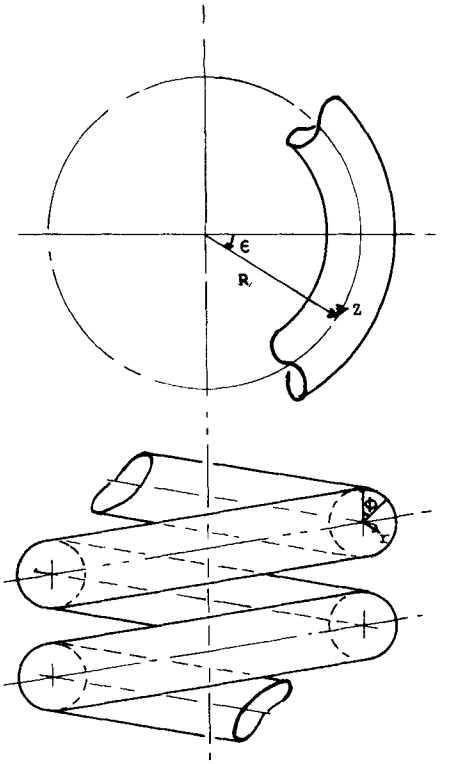

Figure 2. Helical coordinate system used in this work.

$$\frac{\partial\theta}{\partial\phi} = 0 \qquad \text{at } \pm\pi/2 \quad (16)$$

Conditions of an impermeable tube surface leads to

$$\frac{\partial\theta}{\partial\rho} = 0 \qquad \text{at } \rho = 1 \quad (17)$$

Further, we assume that a fluid with constant concentration ($C = C_I$) enters the tube giving

$$\theta = 1 \qquad \text{at } Z = 0 \quad (18)$$

Note that Eqs. 4–15 are valid for fully developed velocity profiles. The likely error due to this assumption will be discussed briefly.

The question of development of velocity profiles in curved tubes has been examined by a number of authors in the case of a Newtonian fluid (Austin and Seader, 1974; Agrawal et al., 1978; Yao and Berger, 1975; Humphry, 1978). They have shown that the hydrodynamic entrance lengths for coiled tubes are relatively short. For instance, for developing curved tube laminar flow with a parabolic velocity profile at the inlet, the experimental data by Austin and Seader (1974) supported by the numerical computations of Patankar et al. (1974) show that the velocity profiles are essentially established within the first quarter-turn of the coil. No prior studies have appeared for power-law fluids due to the enormous complexity of the hydrodynamic problem. However, it is anticipated that the entrance lengths would be even shorter for power-law fluids as is commonly known from the corresponding studies in a straight tube (Mashelkar, 1974).

In general we may note that for chemical reactors $L/D \gg 1$ and the influence of such entrance effects especially for high Schmidt

TABLE 1. LOCAL CONCENTRATION DISTRIBUTION IN A COILED TUBE FOR A NEWTONIAN FLUID

$\xi = 0.0528$; $De = 10$; $Sc = 1{,}000$; $\alpha = 10$; $\theta_{avg} = 0.5972$							
ρ/θ	$-\pi/2$	$-\pi/3$	$-\pi/6$	0	$+\pi/6$	$+\pi/3$	$+\pi/2$
0	0.5259	0.5259	0.5259	0.5259	0.5259	0.5259	0.5259
0.2	0.5186	0.5792	0.6242	0.6379	0.6248	0.5803	0.5287
0.4	0.5214	0.6065	0.6669	0.6864	0.6678	0.6086	0.5348
0.6	0.5157	0.5916	0.6448	0.6565	0.6467	0.5945	0.5348
0.8	0.5014	0.5459	0.5718	0.5810	0.5735	0.5511	0.5322
1.0	0.4911	0.5081	0.5152	0.5165	0.5161	0.5142	0.5121

number systems is negligible, as shown by Mashelkar and Venkatasubramanian (1983) in the case of a straight tube reactor.

Procedure for Numerical Solution

Equation 3 together with the boundary conditions represents a complete mathematical description of the physical system. It was solved by using Brian's (1961) modification of the Peaceman-Rachford (1955) alternating-direction implicit (ADI) method. This method retains all the advantages of implicit solutions and avoids the disadvantages encountered in explicit methods. The details of the numerical algorithm used can be obtained by writing to us.

The details of the concentration field $\theta(\rho,\phi)$ were first obtained. The bulk average concentration was obtained by averaging the local concentration values as

$$\theta_{avg} = \frac{\int_0^1 \int_{-\pi/2}^{\pi/2} \theta \rho w d\rho d\phi}{\int_0^1 \int_{-\pi/2}^{\pi/2} \rho w d\rho d\phi} \tag{19}$$

The accuracy of the numerical solutions was checked by obtaining the concentration field $\theta(\rho,\xi)$ and $\theta_{avg}(\xi)$ in the limiting case of a laminar tubular flow reactor (note that ξ is defined as Z/Sc) and comparing these with the known solutions in the literature. There was a very good agreement between the two solutions.

RESULTS AND DISCUSSION

We shall focus our attention on the following questions:

1. What are the principal qualitative and quantitative differences that the secondary flow has on the details of the concentration field?
2. Reynolds number and radius of curvature influence the secondary flow. What is the effect of these variables on the local concentration field and also the bulk average concentration?

We shall consider each of the above in some detail.

Concentration Field in a Coiled Tube Reactor

It is obvious from Eq. 1 that the local concentration profiles will be functions of both the angular as well as the radial coordinate at a given axial distance. This is unlike the case of a straight tube where, in view of the angular symmetry, the concentration profile is a function only of the radial distance. In Table 1 we have shown some typical concentration profiles in a helical coil for $De = 10$ and $Sc = 1{,}000$. The value of the homogeneous reaction parameter α has been fixed at 10. What is striking is the fact that the bulk average concentration is 0.5972 whereas at none of the points in the coiled tube does the concentration differ by more than 18% from the mean value. This point is more vividly brought out in Figure 3, where the local concentration profiles for a laminar tubular reactor, a plug flow reactor, and a coiled tube reactor have been plotted in a given plane around the vertical axis and the horizontal axis. It will be recalled that in a plug flow reactor, there is no distribution of residence times and the radial diffusivity is infinite. In the case of a laminar tubular flow reactor, there is a considerable axial dispersion effect. It is readily seen that the concentration field in a coiled tube reactor shows a remarkable difference in comparison to that in a tubular reactor. The concentration profile is almost flat on the horizontal axis at least for low values of α. The concentration profile on the vertical axis has two maxima. This is understandable when one examines the nature of the streamlines on the vertical axis (Figure 1). The points marked A and B are the points of maximum axial velocity and therefore have the smallest residence times. The two maxima seen in Figure 3 are related to these maxima.

In Figure 4 concentration profiles in the horizontal plane and the vertical plane are shown for large values of the reaction parameter α. It can be seen that the flatness of the concentration profile which existed in the case of low α values has now disappeared considerably. A possible mechanistic explanation emerges when one considers the role of the convective field which brings in fluid from around the wall to the interior of the core. The speed with which the reaction occurs will determine this flatness. The overall difference between the laminar tubular reactor and the coiled tube reactor can easily be understood when one recalls that reaction rate for a first-order reaction is proportional to the concentration at that point. In a tubular reactor, the fluid elements at

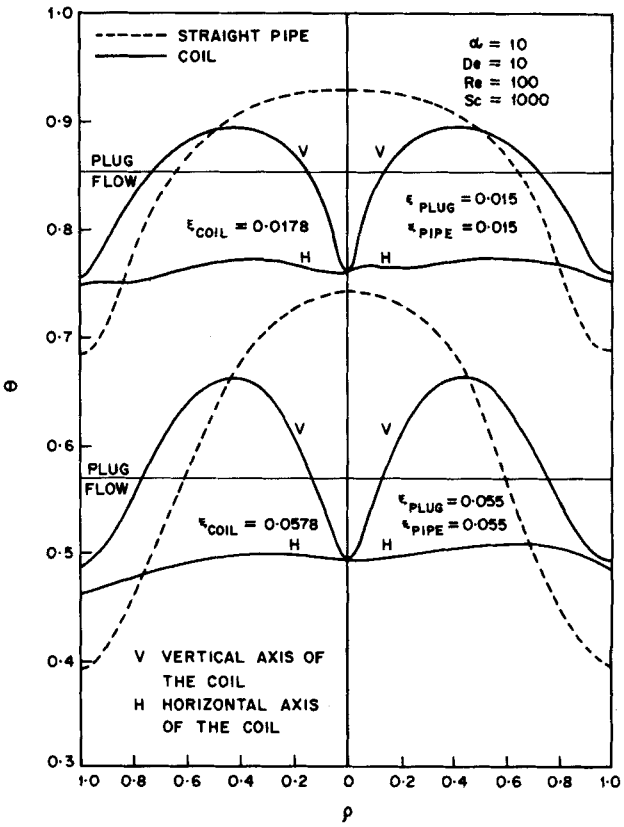

Figure 3. Comparison of the local concentration profiles for laminar tubular, helical coil, and plug flow reactors.

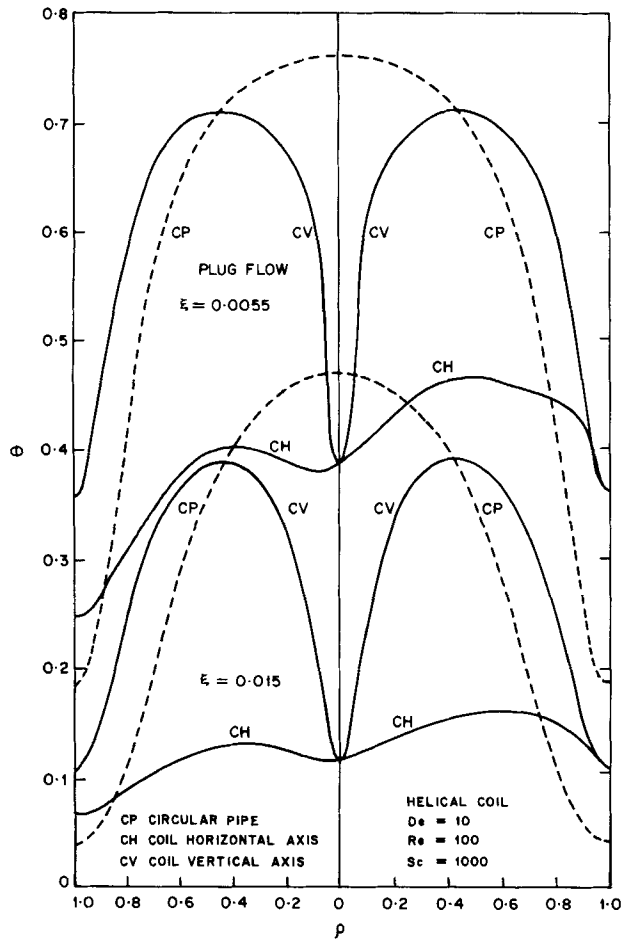

Figure 4. Comparison of the local concentration profiles for laminar tubular, helical coil and plug flow reactors.

different points have different velocities and therefore they undergo reaction to different extents. The elements near the wall undergo more reaction due to smaller residence times, and vice versa for the elements in the central core. The only mechanism by which the elements move across the radial direction is molecular diffusion, which is an extremely slow process. On the other hand,

TABLE 2. INFLUENCE OF REYNOLDS NUMBER AND CURVATURE RATIO ON CONVERSION IN A COILED TUBE REACTOR FOR A NEWTONIAN FLUID

Sc = 1,000; α = 10

	a/R = 0.0225		a/R = 0.01
ξ	Re = 10	Re = 100	Re = 100
0.0008	0.9919	0.9899	0.9915
0.0478	0.6296	0.6252	0.6267
0.0678	0.5202	0.5154	0.5168
0.0828	0.4508	0.4460	0.4474

in a coiled reactor, the finite radial velocity component brings in the elements from nearer the wall and takes away the elements nearer the center to the outer wall. This helps in smoothing out the differences in the residence time distribution and also the extent of axial dispersion, thereby causing considerable smoothing in concentration profiles.

One can see a close approach toward uniformity in concentration flow field even when the intensity of secondary flow under the low Dean number conditions examined here is so mild. This can be explained quantitatively by specific reference to Figure 4. For instance at ξ = 0.015, the ratio of maximum to minimum concentration in a tubular reactor is 12.4; whereas in the case of helical coils it is 2.48 and 3.82 in the horizontal and vertical planes, respectively. Both these are at a fixed values of De = 10, Re = 100, and Sc = 1,000. The profiles in a coiled tube are thus more uniform than those in a straight tube.

Average Concentration in a Coiled Tube Reactor

We shall now examine the influence of Re and the curvature ratio on the cup mixing concentration θ_{avg} or the conversion $1 - \theta_{avg}$. In Table 2 we have presented data to indicate the influence of Re on the bulk average concentration. With an increase in Re and the curvature ratio, there is a marginal improvement in the performance of the reactor. This is explained when reference is made to Eqs. 4 and 5 which show that the secondary velocity components u and v, which are responsible for the cross-sectional mixing, are proportional to both Re and the curvature ratio. We can consider the difference in the bulk average concentration in a coiled and in a straight tube reactor. From the previous discussion, it is evident that the performance obtainable in a coiled reactor

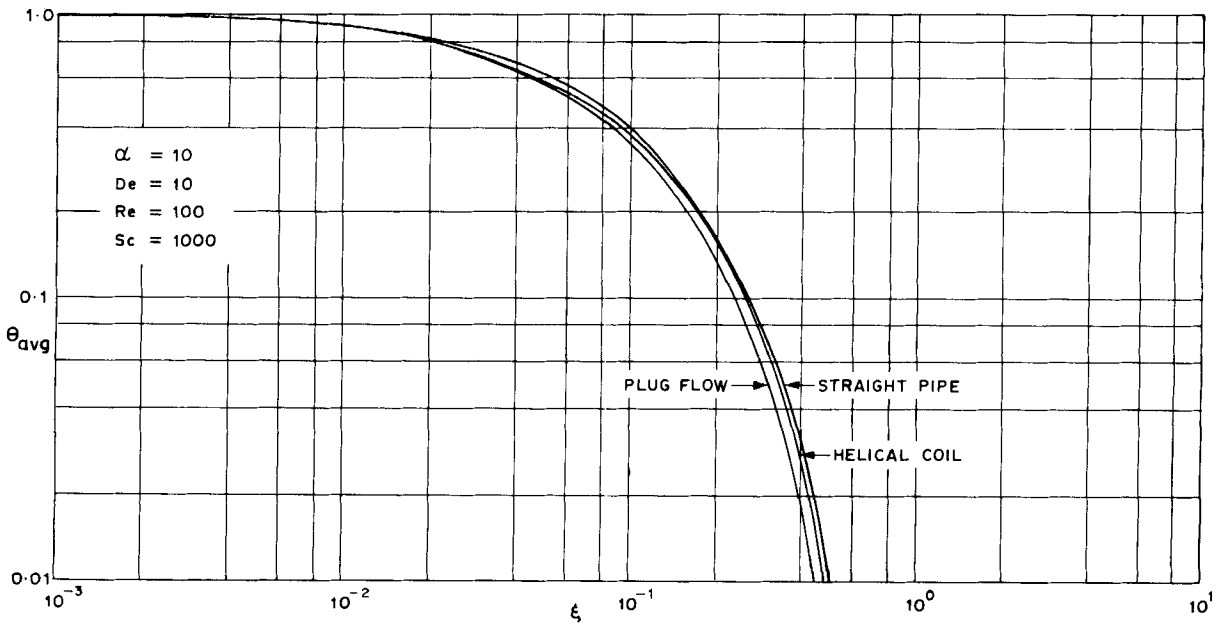

Figure 5. Comparison of the bulk average concentrations for laminar tubular, helical coil, and plug flow reactors; α = 10.

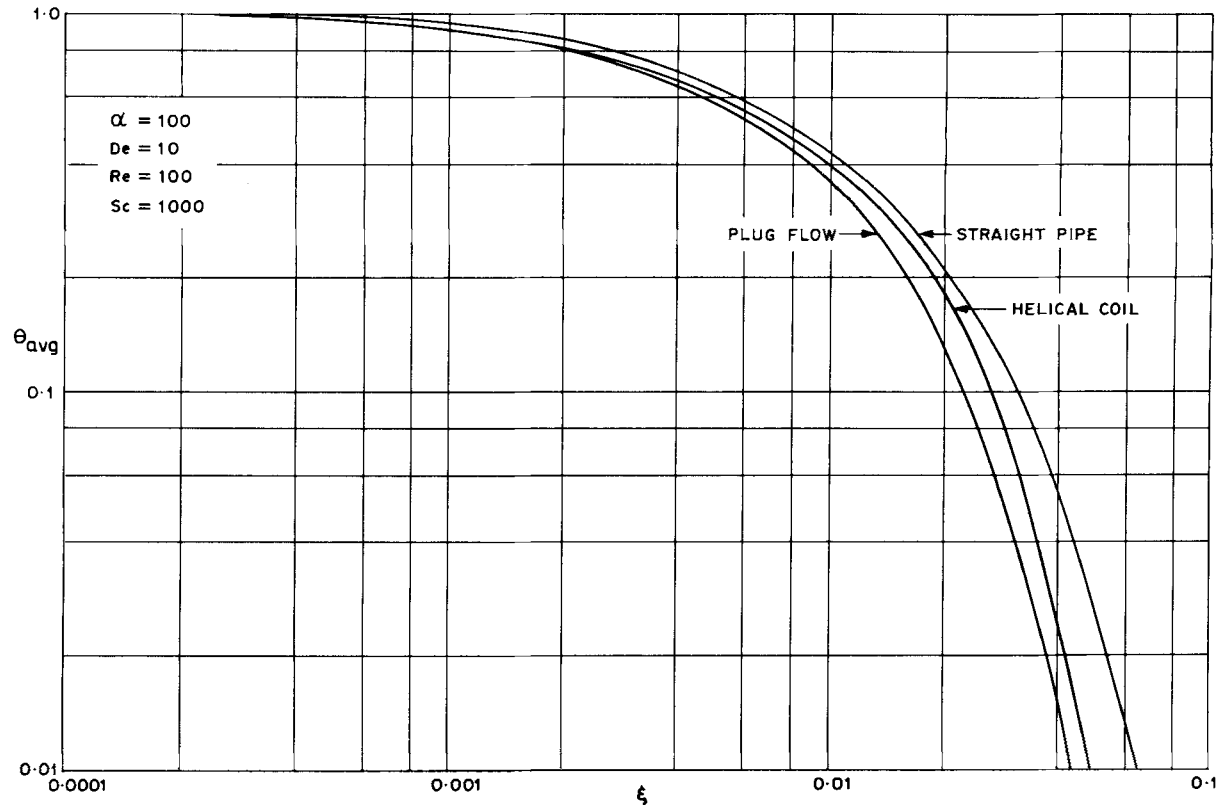

Figure 6. Comparison of the bulk average concentrations for laminar tubular, helical coil, and plug flow reactors; α = 100.

would be between that in the plug flow reactor and that in the laminar flow tubular reactor. Figures 5, 6, and 7 show θ_{avg} as a function of dimensionless axial distance for values of α = 10, 100, and 1,000, respectively. It is indeed seen that the conversion = 1 $-\theta_{avg}$ obtainable in a coiled reactor is higher than that in a straight tube reactor but less than that in a plug flow reactor.

An interesting phenomenon is observed when one considers the influence of different reaction parameters on values of percentage conversions. The comparison can be made by finding out the change in ξ at the same value of θ_{avg}. Smaller ξ for a given θ_{avg} implies smaller holding times resulting in improvement of performance. From Figure 5 it is seen that for α = 10 the improve-

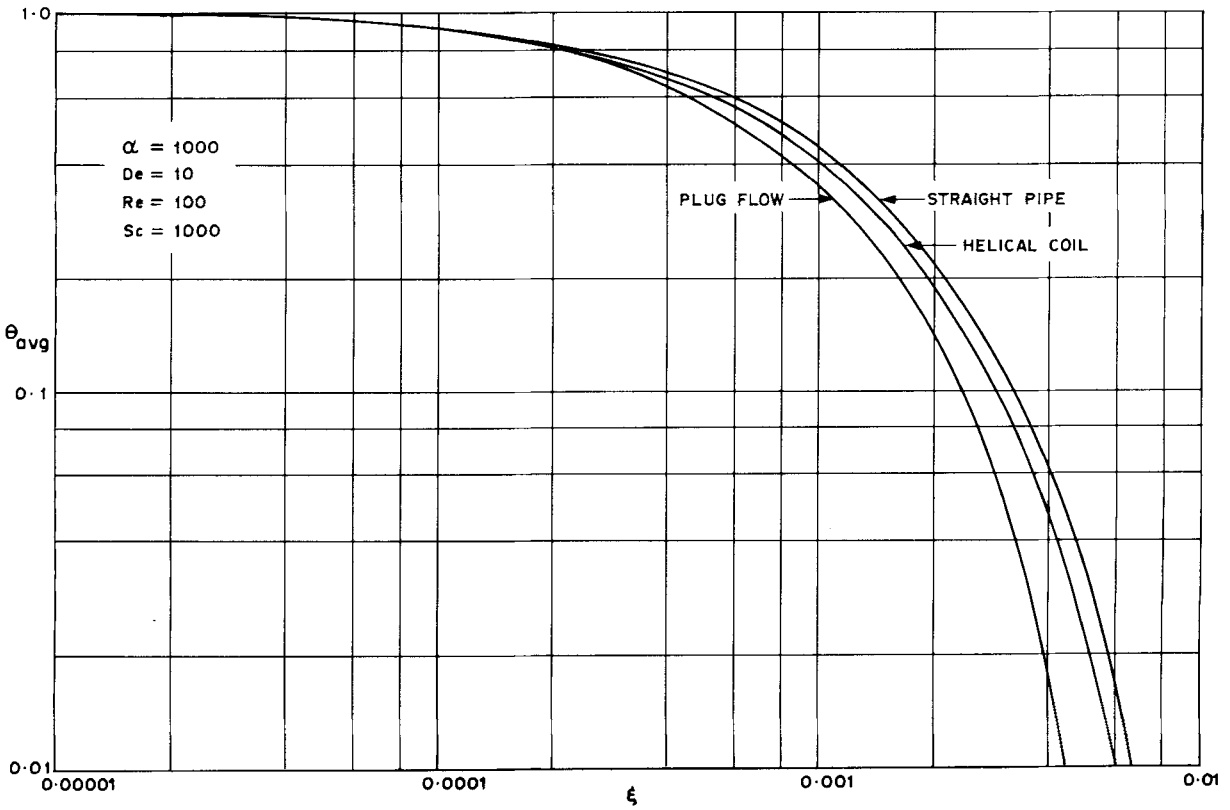

Figure 7. Comparison of the bulk average concentrations for laminar tubular, helical coil, and plug flow reactors; α = 1,000.

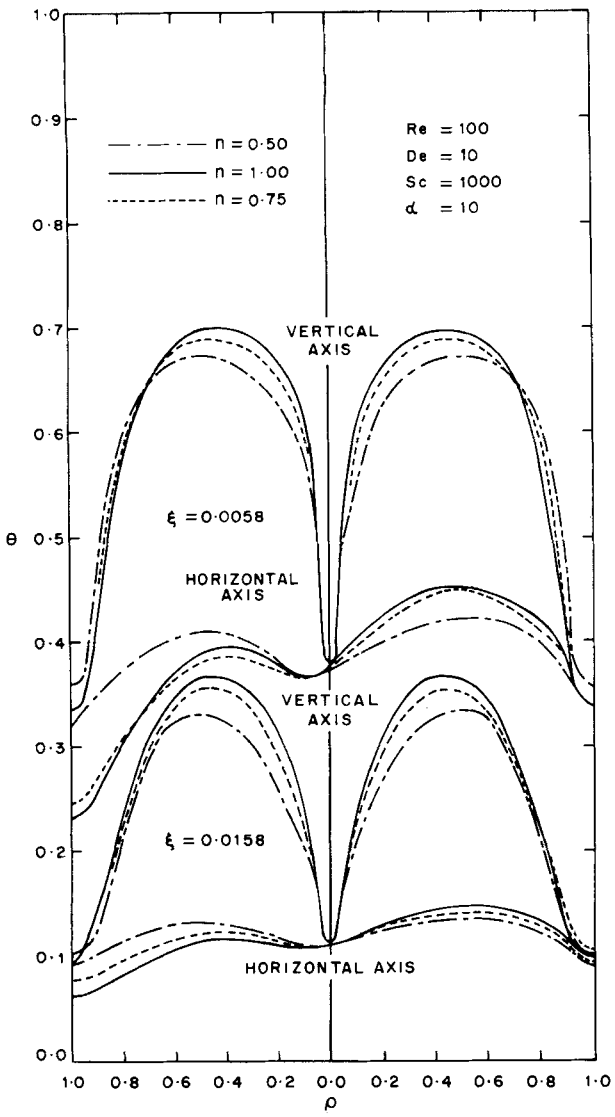

Figure 8. Influence of pseudoplasticity on local concentration profiles.

ment by using a coiled reactor over a laminar tubular flow reactor is about 10% at a conversion level of 80% (i.e., $\theta = 0.20$). For higher reaction rate parameters ($\alpha = 100$), the improvement over a laminar flow tubular reactor at the same conversion level is about 20%. However, as the magnitude of α increases, the improvement over a straight tube reactor appears to diminish.

This phenomenon could be physically explained as follows. There are three characteristic time scales which control the extent of reaction in a straight tube reactor:

$1/k$, representative of the speed of the chemical reaction. Higher $1/k$ implies sharper changes in concentration.

a/D, representative of the speed of the radial molecular diffusion. Lower a^2/D implies rapid destruction of radial concentration gradients created by chemical reaction.

L/U, residence time in the reactor.

Additionally a representative time scale should emerge in a coiled tube on account of the secondary flow. The extra radial and angular velocity components present in the coiled tube help in continually moving the fluid from the wall to the center and vice versa. The time scales for creation of a uniform concentration field radially will typically be of the order of the transverse dimension (radius of the tube) divided by the cross velocity component (the maximum radial velocity component). If these time scales were very high (very low secondary flow), then there would be no effect of secondary flow at all on the chemical reaction. More precisely, if the time scales for chemical reactions are very low (high k and very high α) in comparison to the time scales representative of secondary flows, then the influence of secondary flow also will start diminishing. It is this interesting interaction of kinetic, diffusive, and secondary convection time scales that causes the observed trend.

We shall now consider the performance of a coiled tube reactor for a non-Newtonian power-law fluid. For this purpose, we solved Eq. 1 for $n = 0.75$ and $n = 0.5$ to study the influence of pseudoplasticity.

We shall first examine the influence of pseudoplasticity on the concentration profile. Figure 8 gives the concentration profile for $\alpha = 10$. As the extent of pseudoplasticity increases, the local maxima on the vertical plane tend to become smaller while those on the horizontal plane show an inverted behavior. On the left side of the axis, that is toward the inner wall of the tube, the local concentration increases with increase in pseudoplasticity (reduced n), whereas for the elements near the outer wall, this value increases with increase in pseudoplasticity.

Let us now examine the influence of pseudoplasticity on the overall performance of a coiled tube reactor. Figures 9 and 10 show such curves for $n = 0.5$ and $n = 0.75$. It is seen that whether the fluid is Newtonian or non-Newtonian, there is always an improvement when one switches over from a tubular reactor to a coiled tube reactor. It is useful to inquire whether the improvement is more in the case of a Newtonian fluid or a non-Newtonian fluid. The answer to this question can be found by determining the values of ξ at different values of θ_{avg}. Comparison of Figure 6 (Newtonian fluid) and Figure 9 ($n = 0.5$) illustrates the point clearly. It is seen that for $\alpha = 10$, and at a conversion level of 80% ($\theta_{avg} = 0.2$), the improvement is about 10% over a Newtonian fluid, whereas it is 7.6% for a non-Newtonian fluid with $n = 0.5$. This suggests that the effect of pseudoplasticity is to reduce the extent of improvement in performance over that in a laminar tubular reactor marginally.

The second question pertains to the relative effect of the shear thinning property in a coiled tube reactor as against a tubular flow reactor, since the effect of secondary flow as well as pseudoplasticity individually is to reduce the local concentration variation in a given plane. To illustrate the point consider $\alpha = 10$, $De = 20$, $\xi = 0.15$, $Sc = 1{,}000$.

Computations of θ_{avg} were done for a Newtonian fluid ($n = 1$) and a pseudoplastic liquid ($n = 0.5$) in a straight tube reactor. Then by virtue of the fact that the pseudoplastic liquid produces flatter velocity profiles, one obtained an improvement in conversion by 4.5%. Computations were repeated for a Newtonian fluid ($n = 1$) and a pseudoplastic fluid ($n = 0.5$) in a coiled reactor. Now there is a combined effect of shear thinning and secondary flow in this case. The improvement in conversion in a pseudoplastic fluid is 2%. The above calculations show that the stronger flattening effect of pseudoplasticity on the axial velocity profile dominates over the weaker secondary flow effect in the low Dean number range examined here.

We can now specifically examine if the use of a coiled tube reactor will have any practical significance. This can be readily answered by examining the comparisons provided in Figures 5, 6, 7, 9 and 10. Figure 6, for instance, shows that for reaching 99% conversion the length of the reactor required in the case of a helical coil is 32% lower in comparison to a straight pipe reactor. This is achieved for $De = 10$, $Re = 100$. Considering the increase in the length of the straight pipe reactor, the relative pressure drop in a coiled tube is actually 24% lower, calculated by using the numerical computations for low Dean number provided by Austin and Seader (1973). In other words, the weaker secondary flow (at low De) in a coiled tube configuration does not cause any significant additional pressure drop and yet improves the efficiency of the reactor considerably.

It is useful to recall here the observations by Koutsky and Adler (1964) in the high Dean number region. Their study established that secondary flow in helical tubes is an effective mechanism for inhibiting axial dispersion. When compared at equal power consumption, the helical tubes were found to produce less axial dispersion than straight tubes or packed beds. When compared at

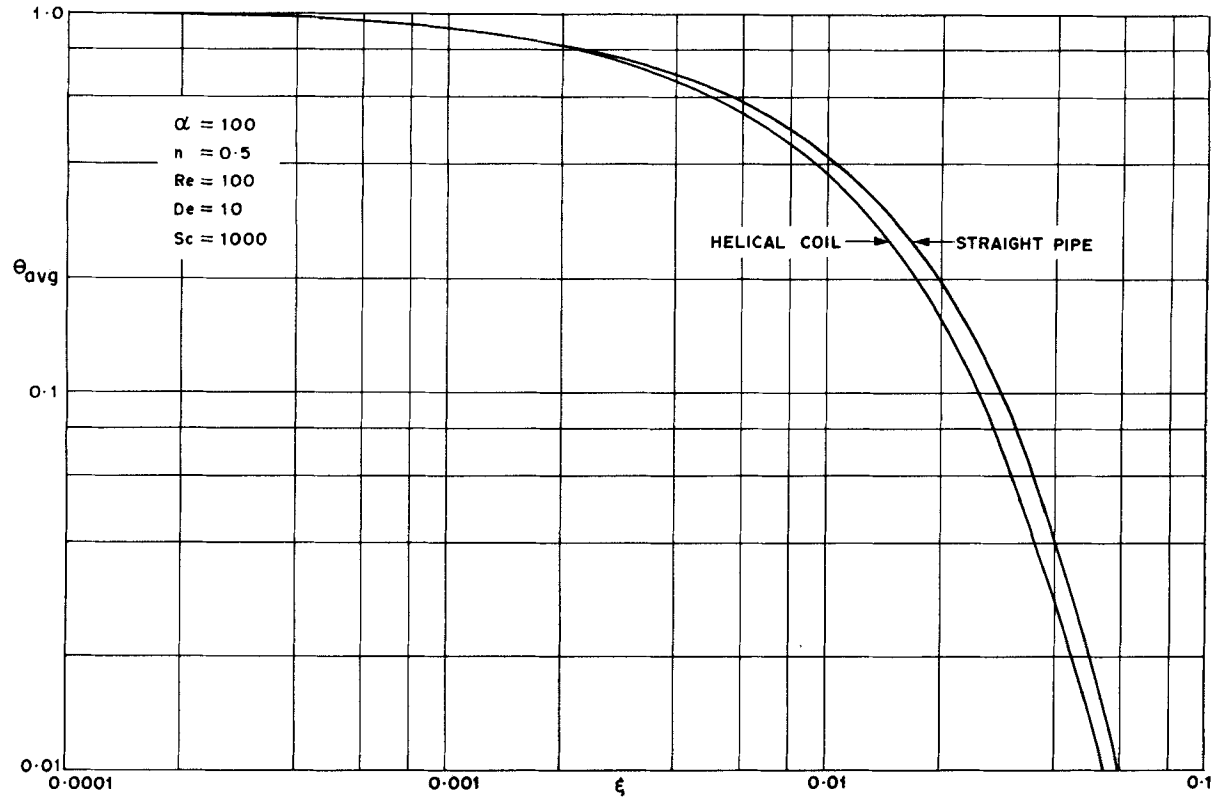

Figure 9. Bulk average concentration profiles for a pseudoplastic liquid; $n = 0.5$.

equal axial dispersion, the helix had less power consumption than the other two systems. For some typical cases of small axial dispersion, the ratio of power consumptions was considerable. Thus the straight tube required about five times the power consumption of the helix and the packed bed about 300 times the power consumption of the helix.

CONCLUDING REMARKS

In the foregoing, we have demonstrated the influence of secondary flow on the detailed concentration field and the average concentration at the outlet in a coiled tube. Even mild secondary flow is shown to smooth out the spatial concentration differences

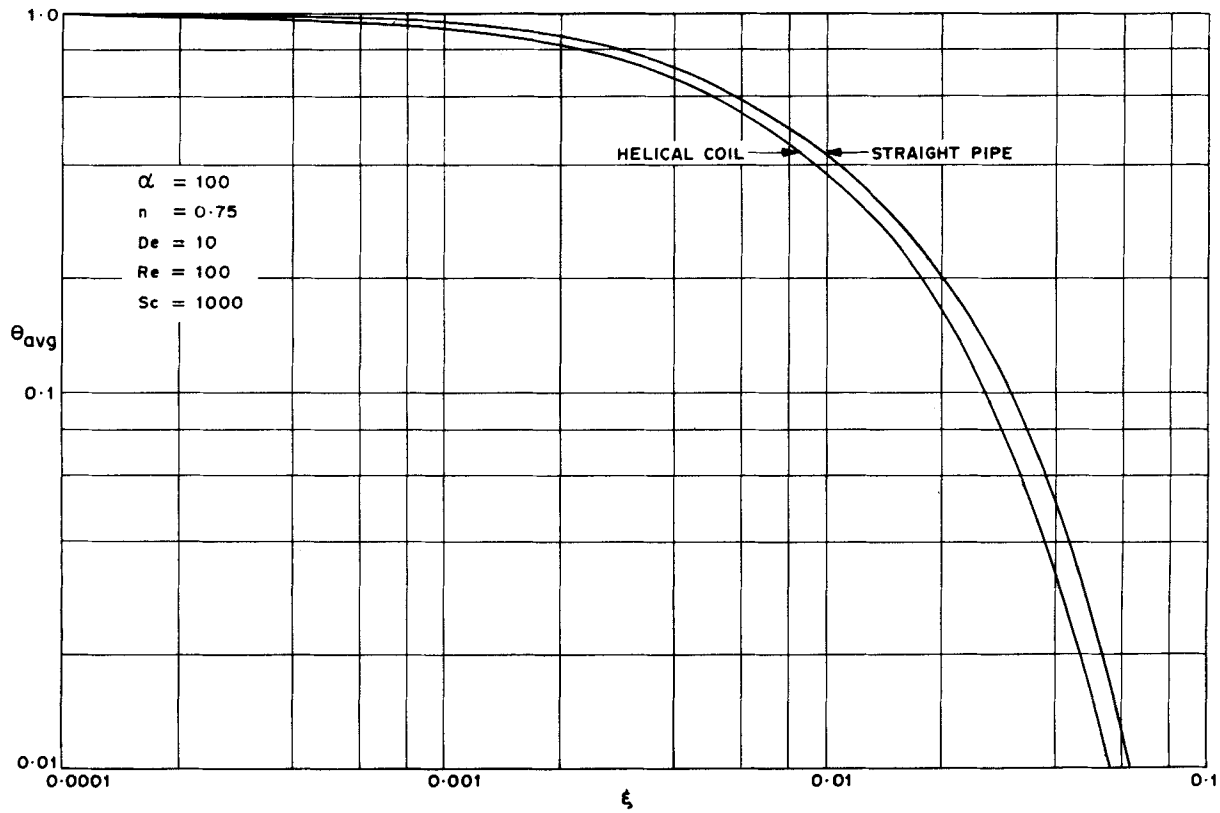

Figure 10. Bulk average concentration profiles for a pseudoplastic liquid; $n = 0.75$.

by a significant amount by smoothing out the residence time distribution or reducing the extent of axial dispersion. The results show strikingly enough that in some parametric range, the advantages in improved conversions can be brought out even by reduction of net power consumption. This may be of great importance while handling complex reaction networks, where selectivity could be a prime consideration. Additionally, such smoothing would imply approach to a more uniform rate of heat generation in exothermic reactions unlike that in a laminar tubular flow reactor. It will be of interest to examine such cases in depth, so that the useful range of applications of a coiled tube reactor can be established.

A special comment may be made concerning the utility of a coiled tube reactor for handling non-Newtonian fluids. Although axial dispersion could be reduced by normally operating the reactor under turbulent flow conditions, it is not desirable to do so in such cases. It is not only due to the obvious disadvantage of enhancement in pressure drop, but also due to the fact that the shear stresses in turbulent flows can be harmful. First, if the medium is polymeric, then the polymer molecules are likely to undergo mechanical degradation in turbulent flow. Second, in case the application in thermal pasturization is considered, then the texture of the liquid food could depend critically on the morphology, which could change very considerably due to the damage caused by high shear stresses under turbulent conditions. The use of secondary flow as demonstrated in this paper becomes particularly relevant in such cases.

The present study was limited to the examination of convective diffusion in a coiled tube under low Dean number conditions ($De < 20$). It will be of interest to investigate the performance of a coiled tube reactor in the high Dean number region, where the influence of secondary flow is considerable.

NOTATION

a = radius of tube
$A(n)$ = function of n, Eq. 9
$B(n)$ = function of n, Eq. 10
C = local concentration
$C(n)$ = function of n, Eq. 11
C_I = inlet concentration
D = diffusivity
$D(n)$ = function of n, Eq. 12
De = Dean number, $Re\sqrt{a}/R$
$f_1(n), f_2(n)$ = functions of n, Eqs. 14 and 15
k = first-order homogeneous reaction rate constant
m = consistency index
n = pseudoplasticity index
r = radial distance
R = radius of curvature
Re = Reynolds number
s = function of n, Eq. 8
Sc = Schmidt number, ν/D
U,V,W = velocity components in r, θ, ψ directions, respectively
u = dimensionless velocity component aU/ν
v = dimensionless velocity component aV/ν
w = dimensionless velocity component W/U_{avg}
U_{avg} = average velocity
w_1 = defined in Eq. 7
Z^* = axial distance
Z = dimensionless axial distance

Greek Letters

α = dimensionless homogeneous reaction parameter ka^2/D
Δ = rate of deformation tensor
θ = dimensionless concentration, C/C_I
ϕ = polar angle
θ_{avg} = bulk-average concentration
ν = kinematic viscosity
ξ = dimensionless axial distance, $Z^*D/U_{avg}a^2$
τ = stress tensor
ρ = dimensionless radial distance
ρ_L = density

LITERATURE CITED

Agrawal, Y., L. Talbot, and K. Gong, "Laser Anemometer Study of Flow Development in Curved Circular Pipes," *J. Fluid Mech.*, **85**, 497 (1978).

Ananthakrishnan, V., W. N. Gill, and A. J. Bardhun, "Laminar Dispersion in Capillaries. Part I: Mathematical Analysis," *AIChE J.*, **11**, 1,063 (1965).

Astarita, G., and R. A. Mashelkar, "Heat and Mass Transfer in Non-Newtonian Fluids," *The Chem. Engr.*, 100, London (1977).

Austin, L. R., and J. D. Seader, "Entry Region for Steady Viscous Flow in Coiled Circular Pipes," *AIChE J.*, **20**, 820 (1974).

Barnes, H. A., and K. Walters, "On the Flow of Viscous and Elastico-Viscous Liquids through Straight and Curved Pipes," *Proc. Roy. Soc.*, **A 314**, 85, London (1969).

Brian, P. L. T., "A Finite Difference Method of High Accuracy for the Solution of Three-Dimensional Transient Heat Conduction Problems," *AIChE J.*, **7**, 367 (1961).

Dean, W. R., "Note on the Motion of Fluid in Curved Pipes," *Phil. Mag.*, **4** (7), 208 (1927).

———, "The Streamline Motion of Fluid in a Curved Pipe," *Phil. Mag.*, **5** (7), 673 (1928).

Greenspan, D., "Secondary Flow in a Curved Tube," *J. Fluid. Mech.*, **57**, 167 (1973).

Homsy, R. V., and R. D. Strohman, "Diffusion and Chemical Reaction in a Tubular Reactor with Non-Newtonian Liquid Flow," *AIChE J.*, **17**, 215 (1971).

Humphry, J. A. C., "Numerical Calculation of Developing Laminar Flow in Pipes of Arbitrary Curvature Radius," *Can. J. Chem. Eng.*, **56**, 151 (1978).

Janssen, L. A. M., "Axial Dispersion in Laminar Flow through Coiled Tubes," *Chem. Eng. Sci.*, **31**, 215 (1976).

Janssen, L. A. M., and C. J. Hoogendoorn, "Laminar Convective Heat Transfer in Helical Coiled Tubes," *Int. J. Heat Mass Transfer*, **21**, 1,197 (1978).

Joseph, B., E. P. Smith, and R. J. Adler, "Numerical Treatment of Laminar Flow in Helically Coiled Tubes of Square Cross Section. Part 1: Stationary Helically Coiled Tubes," *AIChE J.*, **21**, 965 (1975).

Kroutsky, J. A., and R. J. Adler, "Minimisation of Axial Dispersion by Use of Secondary Flow in Helical Tubes," *Can. J. Chem. Eng.*, **42**, 239 (1964).

Katsuri, G., and J. B. Stepanek, "Two-Phase Flow. I: Pressure Drop and Void Fraction Measurements in Co-current Gas-Liquid Flow in a Coil," *Chem. Eng. Sci.*, **27**, 1,871 (1972).

Lynn, S., and J. E. Huff, "Polymerization in a Tubular Reactor," *AIChE J.*, **17**, 475 (1971).

Mashelkar, R. A., "Applicability of Axial Dispersion Model for Non-Newtonian Laminar Flow Tubular Reactors," *Can. J. Chem. Eng.*, **51**, 615 (1973).

———, "Hydrodynamic Entrance Region Flow of Pseudoplastic Fluids: A Simplified Theory," *Proc. Inst. Mech. Eng.*, **188**, 683 (1974).

Mashelkar, R. A., and G. V. Devarajan, "Secondary Flow of Non-Newtonian Fluids. I: Laminar Boundary Layer Flow of a Generalized Non-Newtonian Fluid in a Coiled Tube," *Trans. Inst. Chem. Eng.*, **54**, 100 (1976a).

———, "Secondary Flows of Non-Newtonian Fluids. II: Frictional Losses in Laminar Flow of Purely Viscous and Viscoelastic Fluids through Coiled Tubes," *Trans. Inst. Chem. Eng.*, **54**, 108 (1976b).

———, "Secondary Flows of Non-Newtonian Fluids. III: Turbulent Flow of Viscoinelastic Fluids in Coiled Tubes: A Theoretical Analysis and Experimental Verification," *Trans. Inst. Chem. Eng.*, **55**, 29 (1977).

Mashelkar, R. A., and C. Venkatasubramanian, "Convective Diffusion with Reaction in Developing Flow of a Non-Newtonian Fluid," *Ind. Eng. Chem. Proc. Des. Dev.*, (1983).

McConalogue, D. J., and R. S. Srivastava, "Motion of a Fluid in a Curved Tube," *Proc. Roy. Soc.*, **A307**, 37, London (1968).

Mohan, V., J. Raghuraman, and D. T. Wasan, "Diffusion and Reaction in Turbulent Flow of a Power-Law Fluid in a Confined Tube," *AIChE J.*, **21**, 752 (1975).

Nauman, E. B., "The Residence Time Distribution for Laminar Flow in Helically Coiled Tubes," *Chem. Eng. Sci.*, **32**, 287 (1977).

Nunge, R. J., and B. R. Adams, "Reverse Osmosis in Laminar Flow through Curved Tubes," *Desalination*, **13**, 17 (1973).
Patankar, S. V., V. S. Pratap, and D. B. Spalding, "Prediction of Laminar Flow and Heat Transfer in Helically Coiled Pipes," *J. Fluid Mech.*, **62**, 539 (1974).
Peaceman, D. W., and H. H. Rachford, "The Numerical Solution of Parabolic and Elliptic Differential Equations," *J. Soc. Indust. Appl. Math.*, **3**, 28 (1955).
Raju, K. K., and S. L. Rathna, "Heat Transfer for the Flow of a Power-Law Fluid in a Curved Tube, *J. Indian Inst. Sci.*, **52**, 34 (1970).
Ranade, V. R., and J. J. Ulbrecht, "The Residence Time Distribution for Laminar Flow of Non-Newtonian Liquids through Helical Tubes," *Chem. Eng. Commun.*, **8**, 165 (1981).
Ruthven, D. M., "The Residence Time Distribution for Ideal Laminar Flow in Helical Tubes," *Chem. Eng. Sci.*, **26**, 1,113 (1971).
Seader, J. D., and L. M. Southwick, "Saponification of Ethyl Acetate in Curved Tube Reactors," *Chem. Eng. Comm.*, **9**, 175 (1981).
Srinivasan, P. S., S. S. Nandapurkar, and F. A. Holland, "Pressure Drop and Heat Transfer in Coils," *The Chem. Engr.*, **113**, London (1968).
Trivedi, R. N., and K. Vasudeva, "Axial Dispersion in Laminar Flow in Helical Coils," *Chem. Eng. Sci.*, **30**, 317 (1975).
Weissman, M. H., and L. F. Mockros, "Gas Transfer to Blood Flowing in Coiled Circular Tubes," *Proc. Am. Soc. Civil Engrs. (J. Eng. Mech. Div.)*, **94**, 857 (1968).
Yao, L. S., and S. A. Berger, "Entry Flow in a Curved Pipe," *J. Fluid Mech.*, **67**, 177 (1975).

Manuscript received Aug. 24, 1982; revision received Oct. 20, 1983, and accepted Oct. 20.

A Nonequilibrium Stage Model of Multicomponent Separation Processes

Part I: Model Description and Method of Solution

A nonequilibrium stage model is developed for the simulation of countercurrent multicomponent separation processes. A feature of the model is that the component material and energy balance relations for each phase together with mass and energy transfer rate equations and equilibrium equations for the phase interface are solved to find the actual separation directly. Computations of stage efficiencies are entirely avoided. A procedure for solving the model equations simultaneously using Newton's method is outlined.

R. KRISHNAMURTHY
and
R. TAYLOR
Department of Chemical Engineering
Clarkson University
Potsdam, NY 13676

SCOPE

Rigorous simulation of multistage processes such as distillation or absorption is, more often than not, based upon the equilibrium stage model. This model is well enough known not to need a detailed description here (see, for example, the textbooks by King, 1980, p. 446; Henley and Seader, 1981, pp. 24, 557; Holland, 1975, p. 47; 1981, p. 6). Briefly, of course, the model includes the assumption that the streams leaving any particular stage are in equilibrium with each other. Component *M*aterial balances, the equations of phase *E*quilibrium, *S*ummation equations, and *H*eat balance for each stage (the so-called MESH equations) are solved using one of the very many ingenious algorithms presently available to give product distributions, flow rates, temperatures, and so on.

In actual operation, stages rarely, if ever, operate at equilibrium despite attempts to approach this condition by proper design and choice of operating conditions. The usual way of dealing with departures from equilibrium is by incorporating a stage efficiency into the equilibrium relations. It is with the introduction of this quantity that the problems begin.

The first problem is that there are several different definitions of stage efficiency: Murphree (1925), Hausen (1953), generalized Hausen (Standart, 1965), vaporization (Holland, 1975, p. 268), and others. There is by no means a consensus on which definition is best. Arguments for and against various possibilities are presented by, among others, Standart (1965, 1971), Holland (1975, pp. 268, 327), Holland and McMahon (1970), King (1980, p. 637), and Medina et al. (1978, 1979). Possibly the most soundly based (in a thermodynamic sense), the generalized Hausen efficiencies are ridiculously complicated to calculate; the least soundly based, the Murphree efficiency, is the one most widely used because it is easily combined with the equilibrium equations. Thermal efficiencies may also be defined but almost always are taken to have a value of unity (except, perhaps, when inert species are involved). Whichever definition of stage efficiency is adopted, it must either be specified in advance or calculated from an equation derived by dividing by some reference separation the actual separation obtained from a solution of the component material balance equations for each phase. Many different models of stage efficiency (i.e., different solutions of the component phase balances) have been proposed for binary systems (King, 1980, p. 612). The extension of these models to multicomponent systems provides further complications.

In a c component system there are $c - 1$ independent com-

R. Krishnamurthy is presently with The BOC Group, Murray Hill, NJ 07974.