Linear Eddy Modeling of Reactant Conversion and Selectivity in Turbulent Flows

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The linear eddy model (LEM) is used for statistical predictions of stationary, homogeneous turbulent flows under the influence of isothermal chemical reactions. Nonpremixed reacting systems are considered with two reaction mechanisms: a binary, irreversible single-step reaction $A+B \rightarrow P$; the series-parallel reaction $A+B \rightarrow R$, $A+R \rightarrow P$. In both systems, the influence of various flow parameters on the reactant conversion rate is elucidated. For the second reaction scheme, effects of the flow parameters on the "selectivity" are also investigated. The trends predicted by the LEM agree with those produced previously by direct numerical simulation (DNS) at moderate values of the Reynolds number, Schmidt number and Damköhler number. An important feature of the LEM is its capability to extend the parameter range well beyond that currently attainable by DNS. The LEM generated results for a wide range of Schmidt and Damköhler numbers are discussed as well as their effects on the selectivity. These results assess the performance of some of the existing closures for modeling of the selectivity. None of the closures are capable of reproducing the LEM results.

Introduction

Statistical modeling of turbulent reacting flows continues to be a challenging task (Libby and Williams, 1994). It is well-recognized that one of the stumbling blocks in such modeling is to account for the influence of turbulence on chemical reactions. Due to this difficulty, together with the technological importance of reacting flow devices, significant research has been directed towards understanding the intricate physics of reacting turbulence phenomena. In recent years, the use of direct numerical simulation (DNS) has proven very useful in enhancing our understanding of many important issues in statistical modeling of turbulent reacting flows (Givi, 1989). Due to known limitations of DNS, these efforts have been understandably confined to the analysis of flow phenomena in simple and somewhat idealized configurations. The single-step binary reaction of the type $A+B \rightarrow P$ has been studied extensively by DNS in both homogeneous and nonhomogeneous flows (Givi and McMurtry, 1988; McMurtry and Givi, 1989; Madnia et al., 1992; Frankel et al., 1993; Leonard and Hill, 1988; Riley et al., 1986; McMurtry et al., 1989); see Givi (1994) for a recent review. The results of these studies have provided important information on the mechanisms of reactant conversion in turbulent flows, and have also yielded significant insight in assessing the performance of existing turbulence closures for their predictive capabilities.

More recently, DNS has been utilized for the analysis of more complex, albeit still idealized chemical reaction processes. Gao and O'Brien (1991), Chakrabarti and Hill (1990), and Chakrabarti (1991) have conducted DNS of multistep series/parallel reactions for the purpose of analyzing the evolution of "selectivity" in turbulent flows. The results of these studies are useful for portraying the effects of various flow parameters on the generation of products and in examining the role of important parameters such as the Damköhler number, the molecular Schmidt number, the Reynolds number and stoichiometry on the reactant conversion. The conclusions drawn from these contributions are in general agreement with the accepted trend (Levenspiel, 1972) in that any mechanism promoting mixing tends to yield better selectivity.

Despite the success of DNS in providing significant data and insight on the structure of turbulent reacting flows, there are

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notorious limitations to their applications. Computer requirements limit the application of DNS to a range of flow parameters well below those of practical interest (Givi, 1994). There is clearly a need to extend the parameter range beyond which DNS is able to achieve. In this work a new approach is employed to alleviate some of these difficulties. The approach is based on the linear eddy model (LEM) developed by Kerstein (1988, 1991a). This model portrays some unique features that render it a viable approach for studying turbulent mixing and chemical reaction processes. In particular, the distinction between turbulent stirring and molecular diffusion is maintained at *all* scales of motion. This is achieved by resolving all relevant length scales, even for high values of the flow parameters. This resolution is made computationally tractable by reducing the description of the scalar field to a one-dimensional (linear) spatial domain. In this domain, the effects of physical mechanisms such as advection, molecular diffusion, and chemical reactions are taken into account at all scales. In the context of a one-dimensional description, the physical parameter range that can be considered in LEM is significantly broader than can be currently treated by DNS.

Since its original development, LEM has been used extensively to study various aspects of turbulent mixing and combustion processes. Applications to date include analyses of turbulent reacting shear flows (Kerstein, 1989), turbulent diffusion flames (Kerstein, 1990), turbulent premixed flames (Menon and Kerstein, 1992), mixing in homogeneous turbulent flows (Kerstein, 1991a,b; McMurtry et al., 1993a; Cremer et al., 1994; Frankel, 1993) among others; an up-to-date review is available (McMurtry et al., 1993b). These contributions have been very useful in capturing the physics and in motivating further research on several important features of turbulent reacting flows.

This work uses the LEM to investigate the mechanisms of turbulent reacting flows which are of importance to chemical engineering. For this purpose, the prediction of the reactant conversion in nonpremixed homogeneous turbulent reacting flows is studied considering the same reaction schemes as those used in previous DNS (a single-step reaction mechanism and a series/parallel scheme) (Givi and McMurtry, 1988; McMurtry and Givi, 1989; Gao and O'Brien, 1991; Chakrabarti and Hill, 1990; Chakrabarti, 1991). Model simulations are conducted over a wide range of the flow parameters consisting of the Reynolds number, the Schmidt number, and the Damköhler number(s) to study the effects of these parameters on the statistics of the reacting scalars. The primary intention is the prediction of the chemical conversion for parameter ranges beyond that currently obtainable by DNS.

Linear Eddy Model

Details of the linear eddy model are described in several articles by Kerstein (1988, 1991a,b, 1992); therefore, only a brief description is provided here. The most important feature of the model is its capability to explicitly differentiate among the different physical processes of molecular diffusion, chemical reaction and turbulent convection. This is achieved by a reduced one-dimensional description of the scalar field, which makes it possible to resolve the flow scales even for flows with relatively high values of the Reynolds number, the Schmidt number and the Damköhler number(s). The physical interpretation of the one-dimensional domain is dependent on the particular flow under consideration (Kerstein, 1991a; McMurtry et al., 1993b). In this domain, denoted by x, the evolution of scalar field $\phi(x, t)$ is determined by simulating each of the processes governing its transport. The processes under consideration here involve Fickian diffusion and chemical reaction and are taken into account by the equation:

$$\frac{\partial \phi}{\partial t} = \mathfrak{D} \frac{\partial^2 \phi}{\partial x^2} + \dot{w}_\phi \tag{1}$$

where $\mathfrak{D}$ is the molecular diffusion coefficient and $\dot{w}_\phi$ denotes the chemical source (sink) term. The key to providing an accurate description of turbulent mixing in a one-dimensional formulation is the manner in which the effect of convection is implemented. This is achieved by *random rearrangement events* of the scalar field along the domain. The rules by which these rearrangement processes occur are established such that the random displacements of fluid elements result in a diffusivity that is equal to the "turbulent diffusivity" of the flow. Two parameters govern this process: the frequency of the rearrangement events (stirring) denoted by λ, and the probability density function (PDF) for the size distribution (eddy sizes) of the segments of the flow which are to be rearranged, $f(l)$. To determine explicit expressions for the size and frequency of rearrangement events, a particular rearrangement *mapping* must be chosen. Kerstein (1991a) shows that the *triplet* map portrays several features which suggests its choice for high Reynolds number turbulence simulations. The stirring events induced by this mapping introduce a random walk of a marker particle on the linear domain. Based on high Reynolds number scaling laws, the diffusivity induced by all eddies up to some size l is assumed to scale as $D_T(l) \sim l^p$. The parameter p takes on the value 4/3 for inertial range turbulence, but can be treated as a variable to study other assumed scalings. Based on these scaling arguments, it can be shown that the following relations must be satisfied (Kerstein, 1991a):

$$\frac{L}{\eta} = Re^{1/p} \tag{2}$$

$$f(l) = \frac{3-p}{L\left[\left(\frac{L}{\eta}\right)^{3-p} - 1\right]} \left(\frac{l}{L}\right)^{p-4} \tag{3}$$

$$\lambda = \frac{27}{2} \frac{p}{3-p} \frac{D_T}{L^3} \frac{\left(\frac{L}{\eta}\right)^{3-p} - 1}{1 - \left(\frac{\eta}{L}\right)^p}. \tag{4}$$

Here, L is the integral scale of turbulence, and η is the Kolmogorov length scale. For high-*Re* flows, Kolmogorov scaling relations are obtained for the choice of $p = 4/3$. In this case, it should be noted that LEM is most appropriate for simulating high-*Re* flows since the model is built upon scaling laws which assume just that. However, features of low-*Re* mixing can be studied in a qualitative manner by changing the exponent p in the scaling for the turbulent diffusivity. In particular from Eqs. 3-4, it is noted that an increase of the magnitude of p

Table 1. Conditions for LEM Simulations

Fig.	Sc	Da	Da_1	Da_2	Re	p
1	1.0	-	-	-	30	4/3
2	1.0	-	-	-	30	4/3
3	1.0	∞	-	-	30	4/3
4	-	200	-	-	30	4/3
5	1.0	200	-	-	90	-
6–8	1.0	-	-	-	30	4/3
9	1.0	-	200	20	30	4/3
10	-	-	200	20	30	4/3
11	-	-	20	20	30	4/3
12	1.0	-	200	20	90	-
13	1.0	-	20	200	90	-
14	1.0	-	200	20	30	4/3

results in a lower probability of small-scale stirring events, a larger Kolmogorov scale, and a decrease in the frequency of stirring. Although a quantitative analogy to low-*Re* flows is not provided, these features are consistent with the behavior of low-*Re* flows. It is shown by Cremer et al. (1992) that by increasing the value of p good agreement between the LEM results and moderate-*Re* DNS is obtained. In a more detailed study Cremer et al. (1994) show that an extreme representation of scalar mixing a single length scale l^*, that is:

$$f(l) = \delta(l - l^*) \tag{5}$$

also results in good quantitative agreement with DNS results.

Problem Description and Formulation

The problem under consideration here consists of a homogeneous-isotropic, isothermal, incompressible turbulent flow. The flow field is assumed stationary in that the statistics of the hydrodynamic field are assumed invariant with time. In this way, the influence of turbulence stirring on the chemical reaction process is conveniently captured via the model. Two chemical reaction schemes are considered: A binary single-step reaction of the type:

$$A + B \rightarrow P \tag{6}$$

and the series/parallel reactions:

$$A + B \rightarrow R$$

$$A + R \rightarrow P. \tag{7}$$

The nondimensionalized reaction-diffusion equation for the species concentrations to be solved by the model is given by:

$$\frac{\partial C_i}{\partial t} = \frac{1}{Pe}\frac{\partial^2 C_i}{\partial x^2} + \dot{w}_i \tag{8}$$

Here C_i is the normalized concentration of species i [nondimensionalized with the initial concentration of species A, $(C_A)_0$], t is a normalized time (nondimensionalized with the model eddy turnover time τ_L), and x is the normalized spatial direction (nondimensionalized with the model integral length scale L). With this normalization, Pe is the Peclet number. In this equation, the nondimensional chemical source (sink) terms depend on the nature of the chemistry model. For the scheme given by Eq. 6, we have:

$$\dot{w}_A = \dot{w}_B = -\dot{w}_P = -DaC_AC_B \tag{9}$$

where Da is the Damköhler number defined as the ratio of the turbulent time scale to the chemical time scale. For the series/parallel reaction, Eq. 7:

$$\dot{w}_A = -Da_1C_AC_B - Da_2C_AC_R$$

$$\dot{w}_B = -Da_1C_AC_B$$

$$\dot{w}_R = Da_1C_AC_B - Da_2C_AC_R$$

$$\dot{w}_P = Da_2C_AC_R. \tag{10}$$

Here, Da_1 and Da_2 are the Damköhler numbers for the two reaction steps, respectively. In both cases, the reactants A and B are distributed in an initially segregated manner. This initialization of the scalar field within a domain of size L involves setting $C_A = 1$, $C_B = 0$ for a portion of the domain with size ℓ_A, and $C_A = 0$, $C_B = 1$ for the remaining portion with the size $L - \ell_A$. The magnitude of ℓ_A provides a measure of equivalence ratio. For example, in Eq. 6, $\ell_A = L/2$ yields a stoichiometric mixture. For both reaction schemes, all the other initial concentrations are set to zero.

The configuration considered here portrays some similarities with those treated by DNS (Givi and McMurtry, 1988; Leonard and Hill, 1988; Gao and O'Brien, 1991) and is convenient in analyzing the influence of the flow parameters on the reactant conversion. Here, the primary parameters under consideration are the Reynolds number (*Re*), the molecular Schmidt number (*Sc*), the Damköhler numbers Da, Da_1, Da_2 and the turbulence intensity (through the parameter p). Table 1 provides a listing of the parameter values considered in the simulations.

Results

With the specification of the governing parameters, model simulations are conducted as follows. Along the one-dimensional domain, the effects of molecular diffusion and chemical reaction are taken into account as continuous processes governed by Eq. 1. These equations are solved numerically by means of a standard forward-time, central-space finite difference discretization with periodic boundary conditions. To simulate the influence of turbulence, at discrete time intervals which are determined by the parameter λ, the molecular processes are interrupted by rearrangement events. The size of the rearranged domain is randomly selected from the distribution $f(l)$. This process is continued until a specified time has elapsed. Because stirring is implemented as a random process (subject to rules), the statistical properties of the flow are not guaranteed to be the same for different realizations of the same initial configuration. Statistical properties of the scalar field are therefore obtained by averaging the results over many separate individual realizations. The total resolution is determined by the number of elements discretizing the initial field, and also by the total number of realizations. In the simulations conducted here the total resolution consists of 8×10^4 elements (that is, 200 grid points × 400 realizations) for $Re=30$ and twice that amount for $Re=90$. In each realization, the number of grid points is such that the distance between the neighboring nodes is less than the Batchelor scale (McMurtry et al., 1993a). Sensitivity of the results to temporal and spatial resolution as well as to the number of realizations was checked for convergence. With the assumption of spatial homogeneity along one dimension, the total number of elements also provides a means of constructing the statistics. Such statistics typically consist of the ensemble-mean species concentration, $\langle C_i \rangle$ and the second-order correlations of the type $\langle C_i' C_j' \rangle$. Here, $\langle\ \rangle$ and the prime denote the ensemble-average and the deviation from this average, respectively. For the single-step reaction, the "unmixedness ratio" is defined by the ratio of the normalized second-order moment $\langle C_A' C_B' \rangle$ to that under nonreacting conditions:

$$\mathcal{Z}(t)=\frac{\left.\dfrac{\langle C_A' C_B' \rangle t}{\langle C_A' C_B' \rangle 0}\right|_{Da}}{\left.\dfrac{\langle C_A' C_B' \rangle t}{\langle C_A' C_B' \rangle 0}\right|_{Da=0}}. \tag{11}$$

For the series/parallel scheme, the selectivity is defined as (Bourne, 1982):

$$X_s(t)=\frac{2\langle C_P \rangle}{2\langle C_P \rangle + \langle C_R \rangle} \tag{12}$$

which adopts a value between 0 and 1. Considering R as the "desired" product, the selectivity improves as X_s decreases and perfect selectivity is achieved when $X_s=0$. From a practical standpoint, determination of selectivity is an important issue, because it relates to the efficient usage of raw materials as well as the effort involved in desired product extraction (Levenspiel, 1972).

Results obtained by the model for both reaction schemes are discussed in order. The simulations are conducted on a Cray-YMP computer. A typical LEM simulation requires about 5 min of CPU time. This is rather negligible in comparison to time requirements for DNS [typically of order of tens of hours (Givi, 1989)].

Single-step reaction

In Figures 1 and 2 results are presented on the effects of the Damköhler number on the mean reactant conversion and the unmixedness ratio. These results correspond to the case with the full length scale LEM as described by Eqs. 2-4. As expected, an increase in the magnitude of *Da* results in a faster decay of the mean concentration. This trend continues until the limiting case of $Da \rightarrow \infty$, at which the process of reactant conversion is entirely governed by mixing. In this limit, mixing is characterized by the transport of an appropriately defined conserved scalar, typically termed the Shvab-Zeldovich variable (Williams, 1985). For further comparisons, the analytical result recently proposed by Madnia et al. (1992) and Frankel et al. (1992, 1993) for modeling of the maximum rate of reactant consumption is also considered. This result is based on the amplitude mapping closure (AMC) (Chen et al., 1989), which has proven very useful in probability modeling of reacting flows. Figure 1 shows the extent of agreement between AMC and LEM results. The agreements established between AMC and DNS in this case (Frankel et al., 1993; Madnia et al., 1992) therefore intimates the similarity of the LEM and DNS results for the maximum rate of product formation. The disagreement between LEM and AMC (and thus DNS) is better exhibited by the profiles of the unmixedness ratio presented in Figure 2. This figure shows that this ratio also decreases as the Damköhler number is increased consistent with DNS results (McMurtry and Givi, 1989; Madnia et al., 1992). However, the asymptotic value ($t \rightarrow \infty$) of the ratio, as computed by LEM with $p=4/3$ and with $Da \rightarrow \infty$, does not yield the value $2/\pi$. This limiting value is derived by Kosaly (1987) and Givi and McMurtry (1988) under the assumption that the asymptotic PDF of the Shvab-Zeldovich variable is Gaussian. The asymptotic value obtained here is higher. The AMC enforces asymptotic Gaussian-like behavior and thus results in the theoretical unmixedness ratio of $2/\pi$. In Figure 3, similar results from the

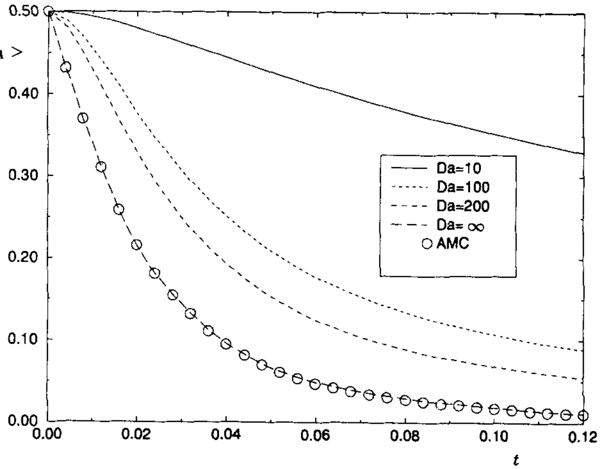

Figure 1. Effect of Damköhler number on reactant conversion for the single-step reaction.

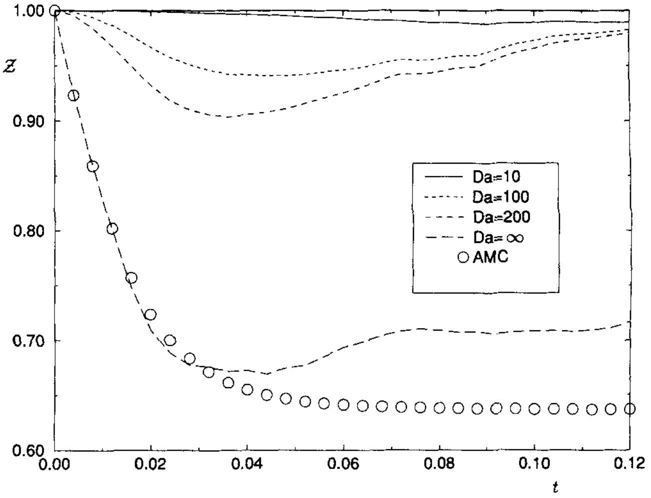

Figure 2. Effect of Damköhler number on the unmixedness ratio for the single-step reaction.

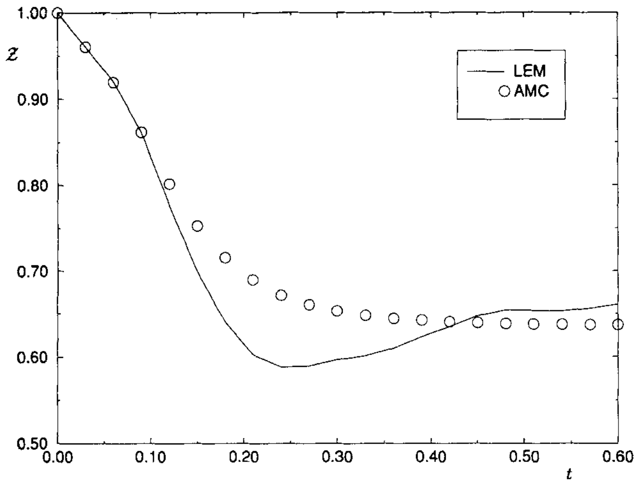

Figure 3. Unmixedness ratio vs. time for the single-step reaction with $Da \rightarrow \infty$.

single length-scale version of the model are presented. Cremer et al. (1994) show that with $l^*/L = 5/12$, this variant of the model is more representative of the mixing processes studied by DNS. In this case, as shown in Figure 3, the asymptotic value of the unmixedness ratio is indeed close to $2/\pi$ and is in better agreement with predictions via AMC and DNS. This feature of LEM has been discussed in detail by McMurtry et al. (1993a) and Cremer et al. (1994). The conclusion is that a "narrow-banded" mixing process, as characterized by a larger value of p, or a single length scale version of the model is more characteristic of the moderate-*Re* mixing of DNS. Therefore, it is at this limit that LEM results are in best agreement with DNS.

In Figure 4, the influence of the molecular Schmidt number on the mean reactant conversion is shown. This figure suggests that at a fixed *Re*, increasing the value of *Sc* leads to a slower mean species decay. This is due to a lower molecular diffusivity leading to less overall mixing at the molecular level. For the isothermal binary reaction scheme, this translates into a slower product formation. The same behavior is observed for the effects of the turbulence intensity as indicated by Figure 5.

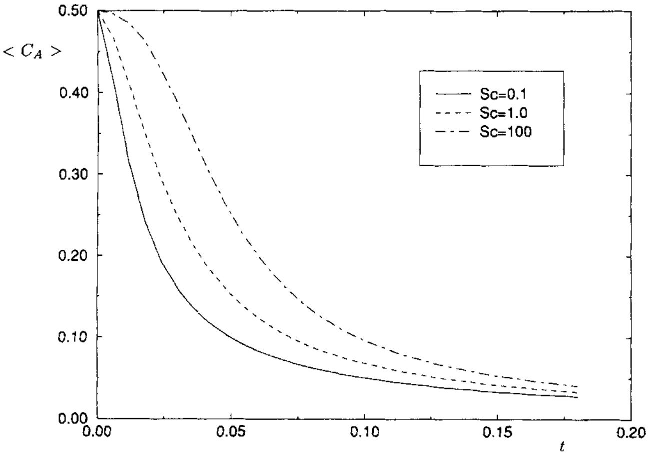

Figure 4. Effect of Schmidt number on reactant conversion for the single-step reaction.

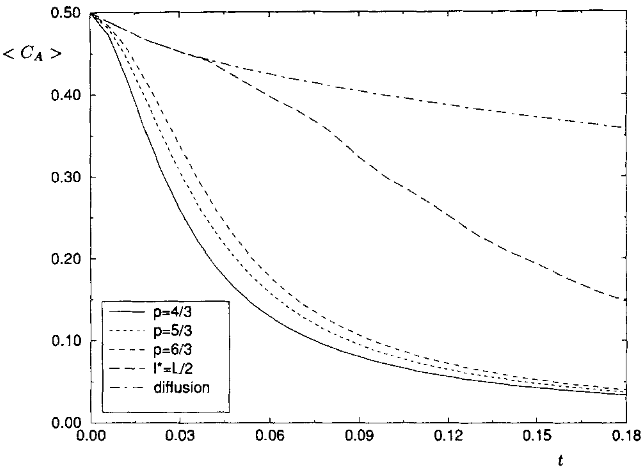

Figure 5. Effect of parameter p on reactant conversion for the single-step reaction.

This effect is mimicked here by varying the exponent p. Also shown in the figure are the results for a single length-scale mixing case and a pure diffusion case. The overall behavior portrayed by this figure is that by decreasing the role of small-scale transport, a slower reactant conversion rate is achieved.

Series/parallel reaction

The influence of the relative reaction rates is studied by varying the magnitudes of the two Damköhler numbers. For Da_1 greater than Da_2 it is expected, and has been corroborated in previous DNS studies (Chakrabarti and Hill, 1990; Chakrabarti, 1991), that more R is produced in the first reaction than is consumed in the second one. This leads to better selectivity. This trend is captured by the LEM as shown in Figure 6. This figure also shows that for a fixed value of Da_2, the selectively is not significantly affected by a change in the magnitude of Da_1. For a fixed value of Da_1, however, there is a substantial change in selectivity when the parameter Da_2 is varied. This trend is in accord with DNS results (Gao and O'Brien, 1991). For a fixed Da_1/Da_2, it is of interest to examine

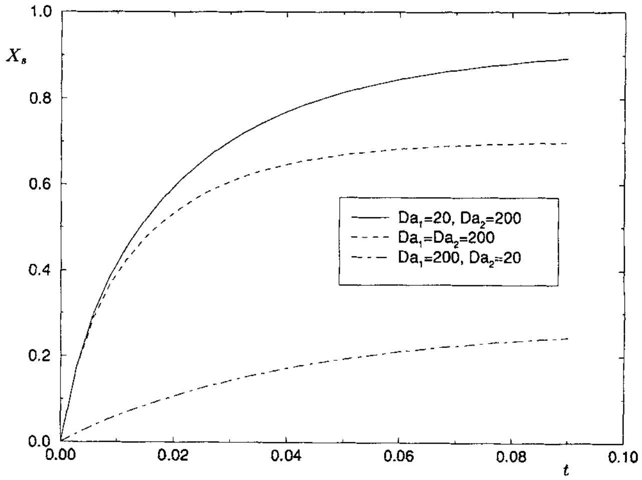

Figure 6. Effect of Damköhler number ratio on selectivity.

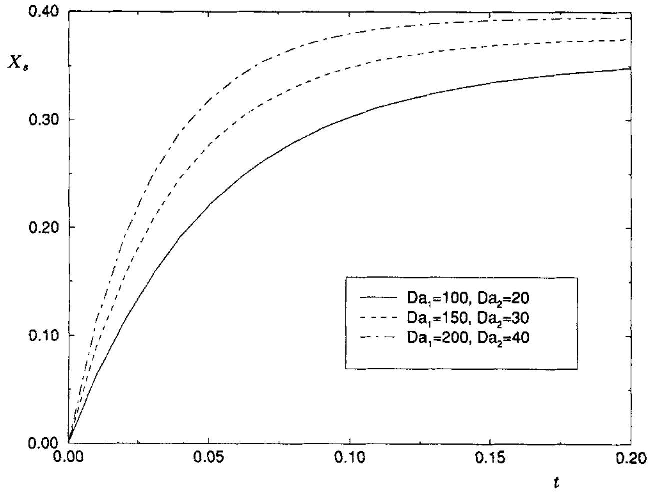

Figure 7. Effect of Damköhler number magnitudes on selectivity.

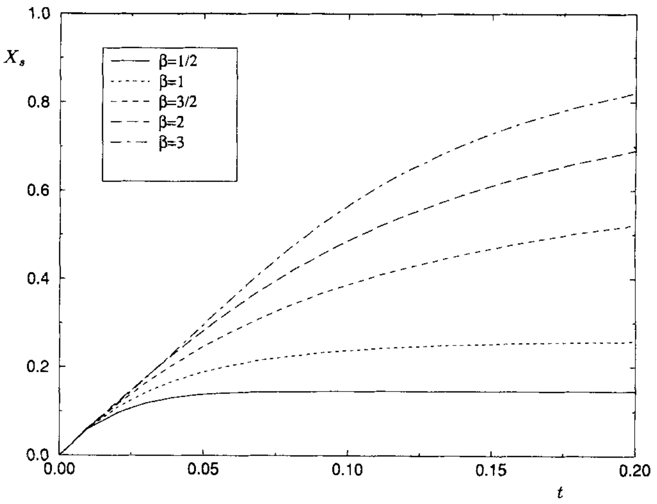

Figure 9. Effect of stoichiometry on selectivity.

the selectivity as the magnitudes of the Damköhler numbers increase. Figures 7 and 8 show the temporal evolution of selectivity for the case where this ratio is greater than unity and less than unity, respectively. In both cases, as the magnitude of Da_1 increases, the selectivity increases. This trend is consistent with that observed in DNS results (Chakrabarti, 1991). From a design standpoint, however, one has little control over this ratio for a given set of reacting species except perhaps by controlling the temperature of the environment.

The effect of stoichiometry, also sometimes referred to as "contacting" (Levenspiel, 1972) is presented in the results of Figure 9. The relative amounts of species A and B injected into the reactor is easily controlled by specifying the initial conditions, as discussed above. The initial stoichiometric ratio, β, is defined as $\beta = \langle C_A \rangle_0 / \langle C_B \rangle_0$. It is observed in Figure 9 that lower values of β lead to better selectivity. This is clear since for $\beta < 1$, the amount of products generated decreases as the magnitude of β is lowered. However, this improved selectivity must be weighed against the fact that associated with lowering β there is a low conversion of B. This results in a waste of raw materials or an increased separation cost.

Figure 10 shows the influence of the Schmidt number on selectivity for the case $Da_1 = 200$, $Da_2 = 20$. For this case the reaction in the first chemistry step is mixing controlled. As the magnitude of the Schmidt number is increased, the fluid mechanics (stirring) becomes relatively more important in the overall mixing process. As a result, the cases with higher values of the Schmidt number yield better selectivity at early times. At later stages of mixing, the scalar gradients are smoothed and turbulent convection is no longer the dominant mixing mechanism. Hence, molecular diffusion dictates the rate of product generation resulting in lower asymptotic values of the selectivity as the magnitude of Sc is decreased. This trend is consistent with that depicted in DNS results of Chakrabarti (1991), albeit at a much narrower Schmidt number range. For the case $Da_1 = Da_2 = 20$, neither of the reaction rates are governed by mixing alone. In this case, the results shown in Figure 11 show the same trend as that in Figure 10 except that the curves do not overlap at the later stages of mixing. This illustrates some of the complex couplings between stirring, dif-

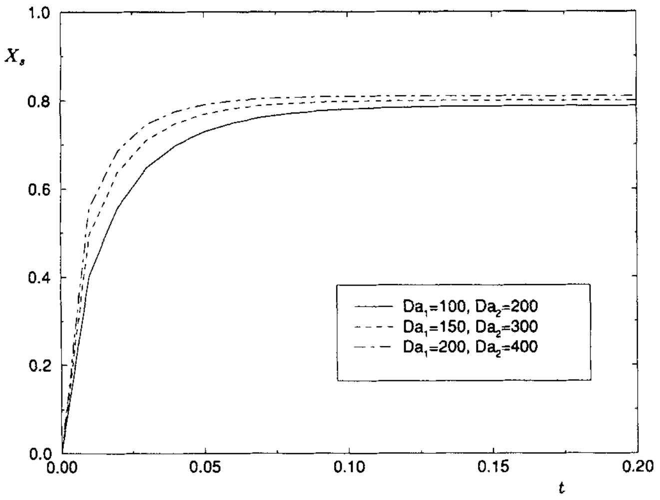

Figure 8. Effect of Damköhler number magnitudes on selectivity.

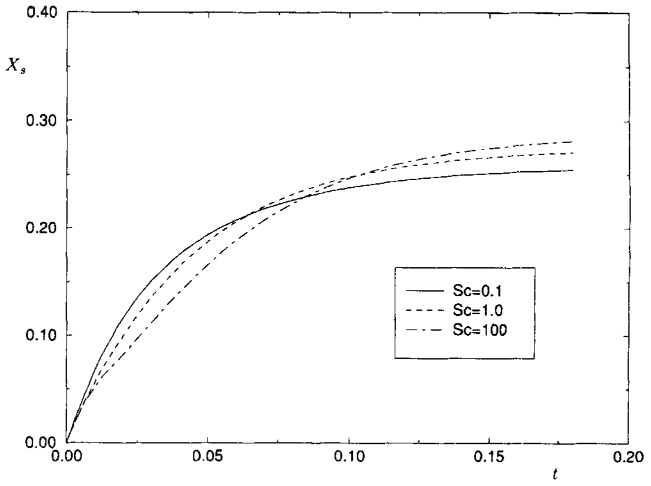

Figure 10. Effect of Schmidt number on selectivity.

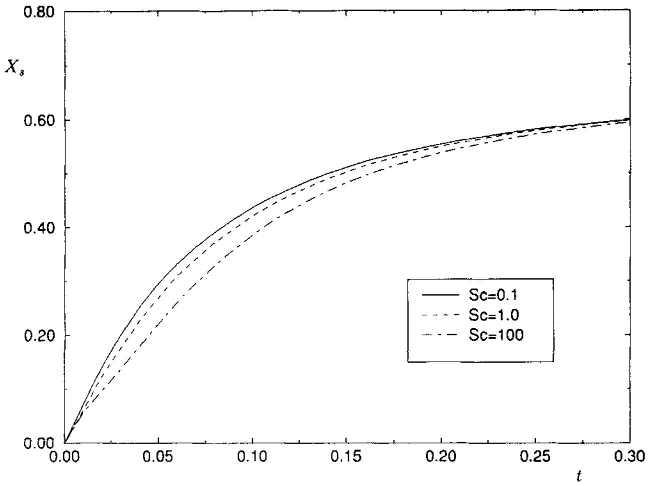

Figure 11. Effect of Schmidt number on selectivity.

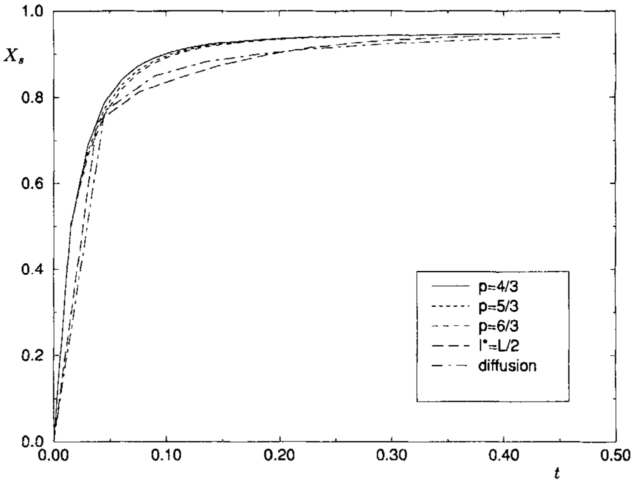

Figure 13. Effect of parameter p on selectivity.

fusion, and reaction for series/parallel reactions. This case has not been considered in any previous DNS; therefore, a direct comparison cannot be made.

The influence of small-scale transport is elucidated in the results presented in Figure 12 for the case where $Da_1 = 200$ and $Da_2 = 20$. Similar to that discussed in the previous subsection, by increasing the magnitude of p, the effects of small-scale stirring become less dominant. Again, for comparison also shown are the results for the single length-scale mixing case and a pure diffusion case. This figure indicates that, consistent with the previous DNS results, the effect of small-scale transport is to improve the selectivity during all stages of mixing and reaction. The effect of turbulence is to smooth the initial scalar gradients through stretching and straining of the scalar field. This leads to more interfacial contact between the reacting species and a more homogeneous scalar distribution. This, in turn, favors production of the desired product and yields better selectivity. These trends are in accord with those portrayed by DNS (Chakrabarti, 1991). However, for the case where $Da_1 = 20$ and $Da_2 = 200$, Figure 13 indicates that by increasing the effectiveness of the small-scale stirring the formation of desired product is not improved. The physical reasoning is that although the second reaction is mixing limited, it is also limited by the generation of product R from the first reaction. For this case, the first reaction is limited by finite rate kinetics. As a result, the parameter p has little effect on the selectivity. Therefore, it can be concluded that turbulence improves selectivity provided that the first reaction is not kinetically limited. This case has not been considered in DNS.

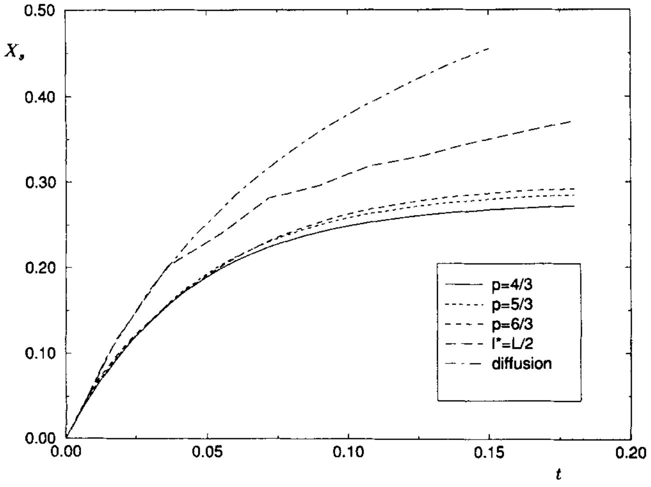

Figure 12. Effect of parameter p on selectivity.

Comparison to other closures

The demonstrated ability of the LEM to reproduce many features of turbulent mixing processes and the noted relationships between the LEM and DNS generated results (McMurtry et al., 1993a; Cremer et al., 1994; Givi, 1989) give credence to the results presented above. As such, a useful application of these results is to guide the development of turbulence models and to assess the performance of existing closures for prediction of the mean reactant conversion rate, particularly for high-Reynolds number flows. In both kinetics schemes considered here, modeling of the scalar covariance has been the issue of primary concern. Reviews of the closures currently available in the literature are given by Heeb and Brodkey (1990) and Chakrabarti (1991). Here the performance of two of the closures are appraised via the LEM. The first model is that proposed by Brodkey and LeWalle (1985) (BL). This closure is based on Toor's hypothesis (Toor, 1962, 1975) and yields the following relation for the covariance:

$$\langle C_A' C_R' \rangle = \langle C_A' C_B' \rangle \frac{\langle C_R \rangle}{\langle C_B \rangle}. \tag{13}$$

The second model is that proposed earlier by Bourne and Toor (1977) and suggests that $\langle C_A' C_R' \rangle$ is approximately zero.

The comparison of the results predicted by LEM with those by the two closures is presented in Figure 14. This figure shows that the LEM results fall between those produced by the other two models and neither closure reproduces LEM results. The trend observed here is similar to that provided by DNS (Chakrabarti, 1991). These results advocate the need for further improvements in future closure strategies in this area. For more practical applications, in a wider range of flow parameters,

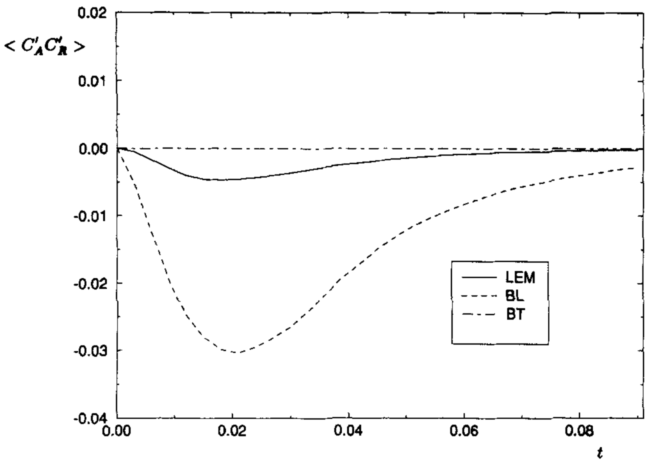

Figure 14. Comparison of the LEM and model predictions for $\langle C'_A C'_R \rangle$.

the LEM can be used to provide detailed information pertaining to the structure of the reacting flowfield and its statistical behavior. In future applications of the model, treatment of more complex chemistry mechanisms is recommended. Also, the model can provide a legitimate substitute for DNS in examining the behavior of other closures in reacting flow systems. Some further speculations and more detailed elaborations on future work in this area are provided by Frankel (1993).

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Notation

A, B, R, P = chemical species
C_i = normalized concentration of ith species, $C_i/(C_A)_0$
D_T = turbulent diffusion coefficient
$f(l)$ = probability density function of l
k_f = speed of reaction
l = size of an eddy
L = integral length scale of turbulence
0 = time zero, inlet of reactor
p = parameter of LEM
Pe = Peclet number, $Pe = Re\ Sc$
Re = Reynolds number, $Re = (L/\eta)^p$
$\dot{w}_i$ = chemical reaction source (sink) term for the species i
x = normalized physical coordinate in the LEM
$\mathcal{Z}$ = unmixedness ratio

Greek letters

λ = frequency parameter of mixing in the LEM
τ_c = chemical reaction time scale, $\tau_c = 1/[k_f(C_A)_0]$
τ_L = Eddy turnover time, $\tau_L = L^2/D_T$
η = Kolmogorov length scale
ϕ = scalar variable

Special symbols

$\langle\ \rangle$ = ensemble average
$'$ = fluctuations from the average

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