Modeling of Isotropic Reacting Turbulence by a Hybrid Mapping-EDQNM Closure

Steven H. Frankel, Tai-Lun Jiang, and Peyman Givi
Dept. of Mechanical and Aerospace Engineering, State University of New York at Buffalo, Buffalo, NY 14260

A hybrid model is developed and implemented for predicting the limiting bound of the reactant conversion rate in an isotropic turbulent flow under the influence of a reaction of the type $A+B \rightarrow$ Products. This model is based on the amplitude mapping closure of Kraichnan for the molecular mixing of a stochastically distributed scalar, and the eddy-damped quasi-normal Markovian (EDQNM) spectral closure for the two-point scalar covariance. The results predicted by this model compare well with available experimental data in both gaseous and aqueous plug-flow reactors, but point to the need for more detailed measurements in future experimental studies. With the implementation of the mapping closure, a simple analytical expression is obtained for the decay rate of the unmixedness. This expression is very convenient and is recommended for direct practical applications in the modeling and design of plug-flow reactors.

Introduction

Statistical modeling of the reactant conversion rate in homogeneous reacting turbulence has been the subject of wide investigations since the early pioneering work of Toor (1962). Among the class of statistical methods in use, it is now accepted that the approach based on the probability density function (PDF) of the scalar quantities is most appropriate (Hawthorne et al., 1949; Toor, 1975; Pope, 1979, 1985; Kollmann, 1990). The principal advantage of this method is based on the fundamental property of the PDF, which includes all the statistical information regarding the reacting field. For this reason, PDF methods have been very attractive and popular as evidenced by their wide use in various forms for the statistical treatment of reacting turbulence phenomena (for recent reviews, see Givi, 1989; Kollmann, 1990; Pope, 1990).

The most systematic means of determining the PDF involves the solution of an appropriate transport equation governing the PDF's evolution. In this equation, due to the nature of the formulation, the effects of chemical reaction appear in a closed form. However, the influences of molecular action cannot be fully described and can be treated only by means of employing an appropriate turbulence closure. In many previous applications, this problem has been addressed through the use of coalescence/dispersion (C/D) models. Examples of such models are the early C/D prototype of Curl (1962) (which was not constructed for turbulence applications, but has been widely utilized for PDF modeling of reacting turbulence), the linear mean square estimation (LMSE) theory of O'Brien (1980), and the closure of Janicka et al. (1979), among others. While not all of these closures were originally presented in a C/D form, it is now established that the majority of those in current use (including the three aforementioned) can be cast in a generalized C/D mold (Pope, 1982; Kosaly and Givi, 1987).

Despite their wide utilization in modeling the transport of scalar variables in turbulence, none of the C/D closures currently in use are physically plausible (Pope, 1982; Kosaly and Givi, 1987; McMurtry and Givi, 1989). In all of these models, the C/D-generated PDFs are not entirely consistent with those either measured experimentally or generated by means of direct numerical simulations (DNS). Specifically, none of these closures are capable of producing an asymptotic Gaussian distribution for the PDF of a conserved scalar from an initially non-Gaussian state in homogeneous turbulence. This trend has been observed in DNS (Eswaran and Pope, 1988; Givi and McMurtry, 1988) and has been corroborated by experimental measurements (Miyawaki et al., 1974; Tavoularis and Corrsin, 1981). This deficiency associated with the C/D closure has been long recognized, and within the past decade significant efforts have been devoted toward developing closures that can

---

Correspondence concerning this article should be addressed to S. H. Frankel.

overcome this nonphysical behavior (for example, Pope, 1982; Norris and Pope, 1991).

For the past few years, Kraichnan and coworkers (Chen et al., 1989) have made rather significant progress in devising a "mapping closure" that can effectively deal with the aforementioned problem. This model, in essence, provides a means of accounting for the transport of the PDF in composition space, and its validity and physical applicability have been convincingly evidenced in a number of comparisons against DNS data (Pope, 1991; Gao, 1991a; Madnia et al., 1991; Jiang et al., 1992). The results of these investigations indicate that, at least for isotropic turbulent transport, this closure is superior to all the previous C/D-type models in depicting a physically plausible PDF evolution.

Based on this demonstrated superiority, we have chosen to utilize the mapping closure for the statistical description of reacting turbulence. For this purpose, we have selected a reacting system under idealized conditions compatible with those considered in many previous works in the chemical engineering community. Namely, the reaction $A+B \rightarrow Products$ in a constant density, isotropic turbulent flow. In this flow, the limiting bound of the reactant conversion rate is predicted by assuming an infinitely fast reaction and ignoring all the nonequilibrium effects associated with the chemical kinetics. In this setting, the only unclosed term requiring a model is the evolution of the appropriate turbulent length scale. This particular closure is not exclusive to the mapping model *per se* and is required in any approach based on a "single-point" statistical description. For this, we have chosen the eddy-damped quasi-normal Markovian (EDQNM) spectral closure. This closure is superior to the commonly used k-ϵ models (Launder and Spalding, 1972), since it includes information regarding the transport of "two-point" statistical quantities. However, its feasibility is currently limited to predictions of flows without spatial inhomogeneities. This does not produce a severe limitation here, since most of the available data on plug-flow reactors display homogeneous characteristics.

In the next section, the problem under consideration is outlined along with the specific assumptions made in developing the hybrid mapping-EDQNM closure. Salient features of the mapping closure at the single-point level are discussed. With this closure, a simple algebraic form is obtained for predicting the limiting bound of reactant conversion. While this algebraic relation is very convenient for practical applications, it is not in a closed form and requires knowledge of the turbulent length scale evolution. The EDQNM closure is capable of providing the required length scale information. In the subsequent section, the formalities of the hybrid closure are discussed, highlighting its relative ease of implementation for practical modeling of plug-flow reactors. And then we compare the results predicted by this combined model with laboratory data for two purposes: (1) to validate the model and (2) to identify some of the relevant parameters in future experiments for such validations. These comparisons are validated by results from a study showing the influence of some factors affecting the rate of reactant conversion. In Section 4, our findings are summarized and some suggestions are made for future work.

Description and Formulation of the Problem

The subject under investigation is a binary reaction of the type $A+B \rightarrow Products$ in an isotropic, isothermal turbulent flow (Toor, 1975; Brodkey, 1975, 1981). The two species A and B are initially segregated and are supplied under stoichiometric conditions. The flow field is assumed constant density, and the influences of the chemical reaction on the dynamics of the turbulence are ignored. The turbulent velocity field is assumed stationary to avoid the complications associated with a varying (decaying) turbulence. This field is parameterized by means of a temporally invariant energy spectrum for the velocity. In this setting, the maximum rate of the reactant conversion is obtained from our hybrid model by implementing the assumption of an infinitely fast chemistry. All the species involved in the reaction are assumed to have identical diffusion coefficients and the same thermodynamical properties. Under these conditions, the statistical behavior of the reacting scalars A, B is related to that of a conserved Shvab-Zeldovich variable, $\mathcal{J}$. This variable is arbitrarily normalized in such a way as to yield $-1 \leq \mathcal{J} \leq 1$. In the framework of the single-point description, the PDFs of the reactants A and B, denoted by $\mathcal{P}_A(\psi, t)$ and $\mathcal{P}_B(\psi, t)$, respectively, are the same for an equivalence ratio of unity. These PDFs are related to that of the Shvab-Zeldovich variable $\mathcal{P}_{\mathcal{J}}(\psi, t)$ by the relation (Toor, 1962; Bilger, 1980):

$$\mathcal{P}_A(\psi,t)=\mathcal{P}_B(\psi,t)=\mathcal{P}_{\mathcal{J}}(\psi,t)+\frac{1}{2}\,\delta(\psi), \qquad (1)$$

where δ is the delta function. Equation 1 demonstrates that all the information pertaining to the statistical behavior of the reacting field is available at all times if the PDF evolution for the conserved scalar field is known. For this, we have employed the mapping closure.

Mapping closure

The implementation of this closure involves a mapping of the random field of interest ψ to a stationary Gaussian reference field η, via a transformation $\psi=\chi(\eta, t)$. Once this relation is established, the PDF of the random variable ψ, $\mathcal{P}(\psi)$, is related to that of a Gaussian distribution $P_G(\eta)$ via (Chen et al., 1989; Pope, 1991):

$$\mathcal{P}(\psi,t)=P_G(\eta)\left(\frac{\partial \chi}{\partial \eta}\right)^{-1}, \quad -\infty \leq \eta \leq +\infty. \qquad (2)$$

In light of this transformation, it is clear that since the random field changes with time, so must the mapping function. Therefore, the probability distribution of the scalar is determined from the knowledge of this mapping function. The transport equation for this function has been developed by Chen et al. (1989). Here, we present only the final results:

$$\frac{\partial \chi}{\partial \tilde{t}}=-\eta\,\frac{\partial \chi}{\partial \eta}+\frac{\partial^2 \chi}{\partial \eta^2}. \qquad (3)$$

In this equation, $\tilde{t}$ is a normalized time within which the scalar length scale information is imbedded. The general solution of this equation has the form (Gao, 1991a; Pope, 1991):

$$\chi(\eta,\tilde{t})=\frac{\sqrt{\Sigma(\tilde{t})^2+1}}{\sqrt{2\pi}\Sigma(\tilde{t})}\int_{-\infty}^{\infty}\chi(\zeta,0)$$
$$\exp\left\{-\frac{[\eta\exp(-\tilde{t})-\zeta]^2[1+\Sigma(\tilde{t})^2]}{2\Sigma(\tilde{t})^2}\right\}d\zeta, \quad (4)$$

where Σ is related to the normalized time by:

$$\Sigma(\tilde{t})^2=\exp(2\tilde{t}-1). \quad (5)$$

For the case of initially segregated reactants, the corresponding forms of the initial PDF and the mapping function are:

$$\mathcal{P}_{\mathcal{J}}(\psi,0)=\frac{1}{2}\,\delta(\psi-1)+\frac{1}{2}\,\delta(\psi+1), \quad (6)$$

$$\chi(\eta,0)=2H(\eta)-1, \quad (7)$$

where H is the Heaviside function. Thus, the solution is obtained by substituting Eq. 7 in Eq. 4 to yield:

$$\chi(\eta,\tilde{t})=\operatorname{erf}\left(\frac{\eta}{\sqrt{2}\Sigma(\tilde{t})}\right), \quad (8)$$

and consequently, from Eq. 2:

$$\mathcal{P}_{\mathcal{J}}[\chi(\eta,\tilde{t}),\tilde{t}]=\frac{\Sigma(\tilde{t})}{2}\exp\left\{-\frac{\eta^2}{2}\,[1-\Sigma(\tilde{t})^{-2}]\right\}. \quad (9)$$

With a combination of Eqs. 1 and 9, all the single-point statistical information regarding the reacting scalar field is determined. A parameter that provides a good measure of the reactant conversion and has been the subject of numerous experimental measurements is the "unmixedness," Ψ^2. This parameter is defined as the normalized fluctuation correlation of the two species (Toor, 1975):

$$\Psi^2=\frac{\langle ab\rangle}{\langle ab\rangle_0}, \quad (10)$$

where $\langle\ \rangle$ denote the ensemble average, and a,b are the concentration fluctuations of the two reactants. The subscript 0 indicates the initial time, that is, at the inlet of the reactor. The temporal evolution of the unmixedness is provided by a combination of Eqs. 1, 9 and 10. After some algebraic manipulations, the final result is in the form (Madhia et al., 1991):

$$\Psi^2(\tilde{t})=\left\{1-\frac{2\arctan[\Sigma(\tilde{t})]}{\pi}\right\}^2. \quad (11)$$

Having such a simple algebraic form for the unmixedness parameter is certainly very pleasing. In the manner presented, however, this equation cannot be compared directly with experimental data. This is due to the form of the final result. The decay of the unmixedness is presented in terms of the normalized time $\tilde{t}$ (which as indicated before contains the length scale information), *not* the physical time t. Note that it is the physical time t that can be translated to the physical axis of the reactor, x, for comparison against actual measurements. In the context of single-point analysis, the relation between this time and the normalized time $\tilde{t}$ cannot be described. This demonstrates the need for some external means by which the knowledge of two-point statistics can be brought into the analysis. To provide this information, we have used the EDQNM spectral closure.

EDQNM closure

The formulation presented in the previous section would be in a closed form with the knowledge of the parameter Σ. Making the assumption that the evolution of the length scales is the same under reacting and nonreacting conditions, the knowledge of the variance of the Shvab-Zeldovich variable, $\mathcal{J}$, would be sufficient to cast Eq. 11 in a closed form. To account for this length scale, the model employed in this work is based on a two-point closure, namely the EDQNM model. This model is a "single-time" (but still two-point) closure, and compared with other satisfactory stationary spectral closures, its computational treatment is relatively less demanding (see Herring et al., 1982, for a review of all spectral closures currently in use). Because of this property, the EDQNM has proven very powerful in turbulence modeling and has been widely utilized in many investigations (see Orszag, 1977; Herring et al., 1982; Lesieur, 1990, for review).

The form of the EDQNM closure in use here is similar to those of Larcheveque and Lesieur (1981) and Eswaran and O'Brien (1989). This is in the form of a physical-space diffusion equation for the two-point scalar covariance in spherical coordinates with an effective diffusivity, that is, laminar plus turbulent. This form is relatively simple in appearance, but still contains most of the essential physics of the Navier-Stokes equations. For the readers who are not familiar with the model, we describe the derivation of the final physical space transport equation for the single-time, two-point scalar covariance. The mathematical derivation for this equation is detailed by Larcheveque and Lesieur (1981) and Lesieur (1991). The derivation of the model begins with the equation for the two-point covariance in the spectral domain as well as the evolution equation for the third-order correlation. The quasi-normal (QN) approximation is based on Millionshtchikov's (1941) hypothesis and is invoked to facilitate solution of the third-order correlation in terms of the two-point covariance, thereby closing the hierarchy of moment equations. The eddy-damping feature provides a model for the effects of the discarded fourth-order cumulants, and the Markovianization allows for a relaxation toward quasi-equilibrium by nonlinear transfers. The resulting integrodifferential equation is in the spectral (wave number) domain and is computationally intensive due to the triadic interactions that arise from the nonlinearities in the transport equations. By making assumptions on the form of the characteristic relaxation time, Larcheveque and Lesieur (1981) were able to transform the covariance equation back into physical space and obtain a version more suitable for computation. This physical space equation in the form utilized is:

$$\frac{\partial}{\partial t}\,\rho(r,t)=\frac{1}{r^2}\frac{\partial}{\partial r}\left\{[\mathcal{K}(r,t)+2\mathfrak{D}]r^2\,\frac{\partial}{\partial r}\,\rho(r,t)\right\}, \quad (12)$$

where the scalar covariance $\rho(r, t)$ is spherically symmetric, $\rho(r, t)=\rho(|r|,t)=\rho(\underline{r},t)$, and is defined as:

$$\rho(\underline{r},t) = \langle \mathcal{J}(\underline{x},t)\mathcal{J}(\underline{x}+\underline{r},t)\rangle. \tag{13}$$

In Eq. 12, the molecular diffusivity is $\mathcal{D}$, and the turbulent eddy diffusivity, denoted by $\mathcal{K}$, is defined by:

$$\mathcal{K}(r,t) = \sqrt{\pi}\int_0^\infty \Theta_\kappa(t)E(\kappa,t)\left[\frac{4}{3\sqrt{\pi}} - \left(\frac{2}{\kappa r}\right)^{\frac{3}{2}} J_{3/2}(\kappa r)\right] d\kappa, \tag{14}$$

where $J_{3/2}$ is the Bessel function of order 3/2, and κ denotes the wave number in the spectral domain.

The eddy diffusivity is determined by the turbulent kinetic energy spectrum function $E(\kappa, t)$. An appropriate EDQNM transport equation can also be constructed for this spectrum by a similar procedure as outlined above. In this work, however, it is assumed that the turbulence field is stationary; therefore, this spectrum is temporally invariant, $E = E(\kappa)$. Finally, the quantity $\Theta_\kappa(t)$, which provides a measure of the characteristic time for triad interactions, is modeled as:

$$\Theta_\kappa(t) = \frac{1 - \exp(-[\mu''(\kappa) + \nu\kappa^2]t)}{\mu''(\kappa) + \nu\kappa^2}. \tag{15}$$

Here, ν is the kinematic viscosity, and the term $\mu''(\kappa)$ is the eddy relaxation frequency which is a measure of the straining effect of scales larger than κ^{-1} on mode $\underline{\kappa}$. This term is modeled as (Larcheveque and Lesieur, 1981):

$$\mu''(\kappa) = \lambda''\left[\int_0^\kappa p^2 E(p)dp\right], \tag{16}$$

where λ'' is a constant taken as 1.3 (Herring et al., 1982).

Model Implementation and Comparison with Laboratory Data

With a combination of these two models, the closure for the statistical variations of the scalar field is completed. The mapping closure provides an analytic expression for the decay rate of the unmixedness, and the EDQNM complements this relation with an estimate of the real-time evolution for the variance of the Shvab-Zeldovich variable. It is assumed that the evolution of the length scales of the Shvab-Zeldovich variable is not affected by the presence of the chemical reaction. This is a reasonable assumption in view of the recent findings of Jiang (1990) and Gao (1990). With this model, the evaluation of the statistical quantities, at least up to second-order statistics (including the unmixedness), is straightforward. However, the implementation of the EDQNM requires numerical integration. Here, the solution of Eq. 12 is obtained by the Crank-Nicolson finite difference scheme, and the numerical integration for the relaxation time and the eddy damping rate (the righthand side of Eqs. 14 and 16) is accomplished using the Clenshaw-Curtis numerical quadrature (Engels, 1980).

In comparing the results generated by this hybrid model to those of laboratory measurements, the information regarding the parameters describing the turbulent field is necessary. The values of these parameters depend on the particular turbulent flow under consideration and must be specified *a priori*. This requirement presents a difficulty when making detailed quantitative comparisons with experimental data, simply because of an apparent lack of extensive measurements in reacting turbulent flows. Therefore, it is necessary in our comparisons with the available data to make some assumptions with regard to some of these unknown parameters.

At the level of the formulation considered here, specific parameters that characterize the turbulence structure are: the kinetic energy spectrum of the turbulence, $E(\kappa)$; the initial shape of the scalar covariance, $\rho(r,0)$; the jet Reynolds number, Re_D; the molecular Schmidt number, Sc; the relative turbulence intensity, I; the integral length scales of both the velocity field, L; the scalar field, $L_{\mathcal{J}}$; and the ratio of the Taylor microscale of the scalar field, $\lambda_{\mathcal{J}}$, to that of the hydrodynamic field, λ. The magnitude of the hydrodynamic integral scale is coupled with the energy spectrum, and the initial size of the scalar integral scale is determined through the specification of the initial distribution of the scalar covariance. The value of the molecular Schmidt number depends on the reacting system under study, and magnitudes of the Taylor microscales are determined by means of isotropic turbulence scaling relations (Tennekes and Lumley, 1972; Brodkey, 1975). Along with these relations, all the other parameters must be specified *a priori*. The assumption of stationary flow precludes any time evolution of the energy spectrum of turbulence, but its spectral distribution must be prescribed. Here, the spectrum is assumed to have the form:

$$E(\kappa) = \begin{cases} C_1\kappa^4\exp\left[-2\left(\dfrac{\kappa}{\kappa_1}\right)^2\right] & \text{for } 0 \le \kappa \le \kappa_1 \\ C_2\kappa^{-\gamma} & \text{for } \kappa_1 \le \kappa \le \kappa_2 \\ 0 & \text{otherwise.} \end{cases} \tag{17}$$

The unknowns C_1, C_2, γ, κ_1, κ_2 are determined from the consistency conditions as described by Jiang (1990). The initial profile of the scalar covariance is assumed to have the distribution:

$$\rho(r,0) = \exp\left[-\left(\frac{r^2}{\lambda_{\mathcal{J}}^2|_0} + C_3\frac{r^4}{\lambda_{\mathcal{J}}^4|_0}\right)\right], \tag{18}$$

where $\lambda_{\mathcal{J}}|_0$ is the initial scalar microscale and C_3 is a constant chosen to be 1.6.

The remaining parameters are specified with the aid of data provided by laboratory measurements. The data considered here are those of Ajmera et al. (1976) and reviewed by Toor (1975). These data are selected since they include information for both gaseous and aqueous reacting systems. Moreover, the reacting fields considered in these experiments are compatible with the assumptions made in our model: very fast reaction involving initially unpremixed, dilute stoichiometric reactants in a constant density, isothermal homogeneous turbulent flow. The plug reactor used in these experiments consisted of a mixing device followed by a reaction tube. The mixing device is a concentric tube in which the reactants are introduced separately in the inner and outer streams. The reacting fields considered are the gaseous nitric oxide and ozone ($Sc = 0.73$), and the aqueous hydrochloric acid and sodium hydroxide ($Sc = 700$). In both of these systems, reactant conversion data are available for cases with jet Reynolds numbers of 3,500, 6,700 and 12,000. There is no information provided on any

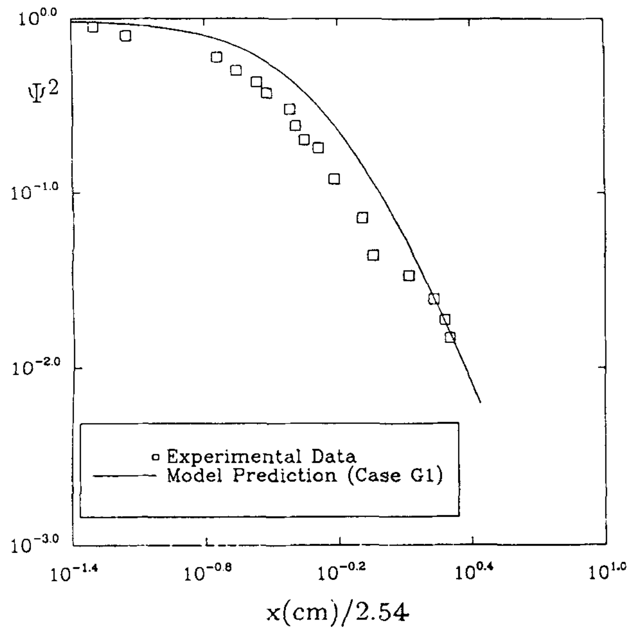

Figure 1. Reactant conversion distance in gaseous system for *Re* = 3,500.

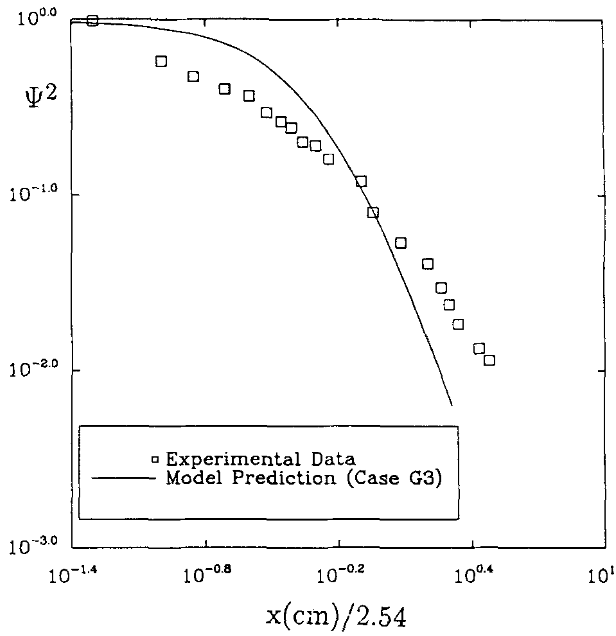

Figure 3. Reactant conversion distance in gaseous system for *Re* = 12,000.

turbulent length scales or the turbulent intensities in these experiments. For comparison, we assume that the reactor tube diameter, D, is representative of the integral scale of turbulence, L, considering a value for the turbulence intensity, I. (The ramifications of these assumptions will be examined later.)

The final results of our predictions are compared with experimental data in Figures 1-6. In these figures, the reactant conversion (Ψ^2)-distance curves for both gaseous and aqueous reactions are presented for all three Reynolds numbers. The relative turbulence intensity in these comparisons was selected as $I = 5\%$, since this value provided the best overall match with the data. This intensity level is perhaps somewhat lower than that of typical fully-developed, homogeneous turbulent flows. In most laboratory flows, however, the turbulence field is usually of decaying nature. Therefore, while at the initial stages of development the laboratory flow may have a higher turbulence intensity, the magnitude of this intensity decreases further downstream. In our stationary turbulence simulations, a constant turbulence level is a compromise and, in essence, represents an average of the corresponding data in laboratory

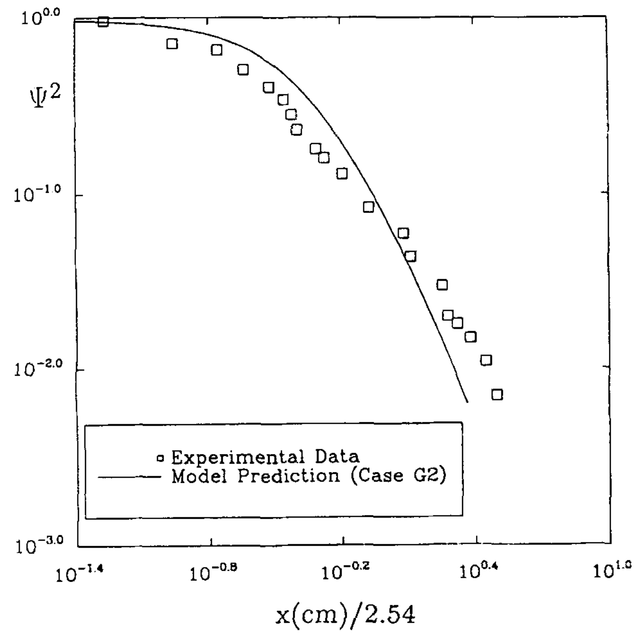

Figure 2. Reactant conversion distance in gaseous system for *Re* = 6,700.

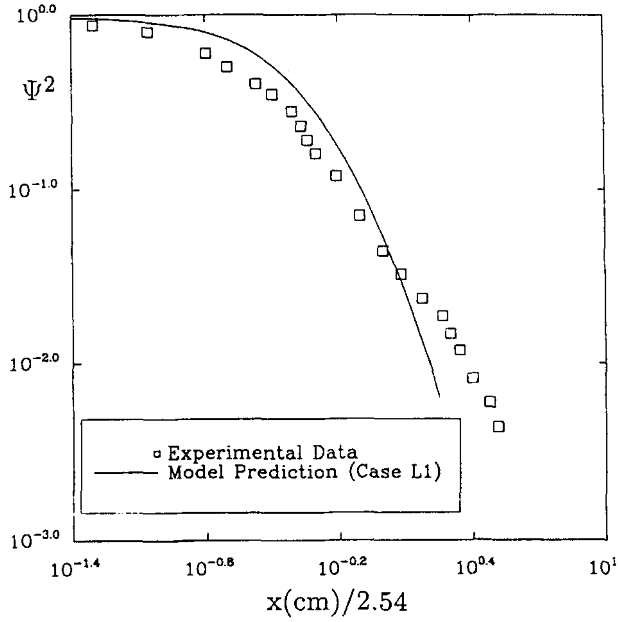

Figure 4. Reactant conversion distance in aqueous system for *Re* = 3,500.

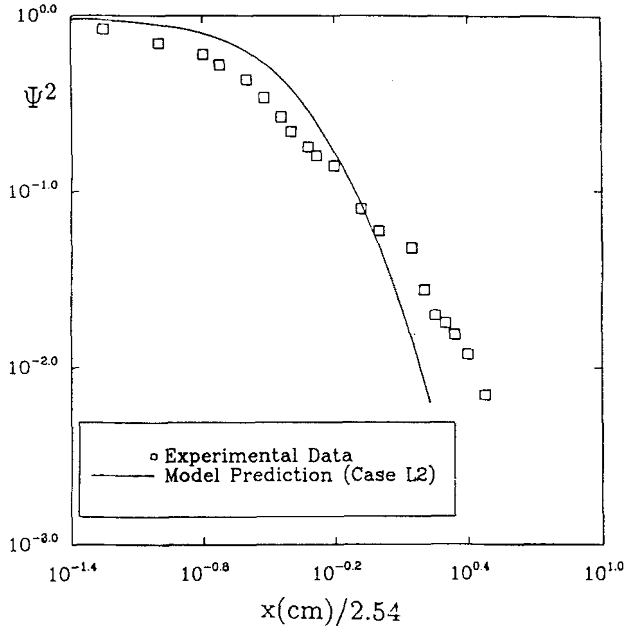

Figure 5. Reactant conversion distance in aqueous system for *Re* = 6,700.

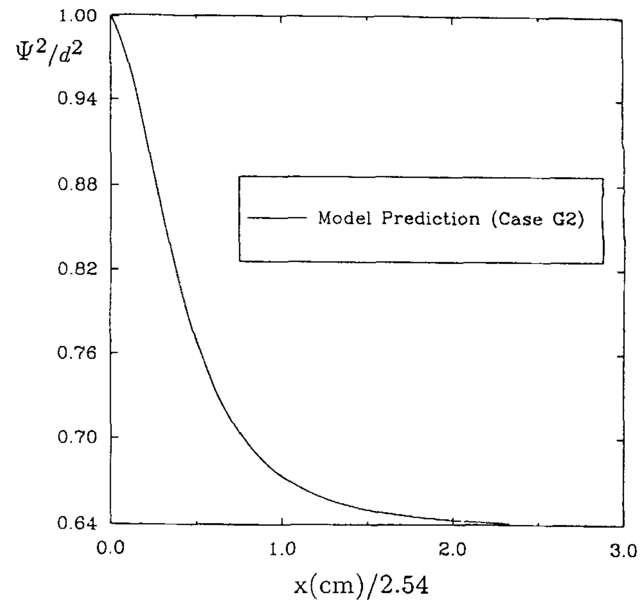

Figure 7. Ratio of reacting to nonreacting unmixedness in gaseous system for *Re* = 6,700.

experiments. The conditions in these figures are identified by the symbols $G1$, $G2$, $G3$, $L1$, $L2$, and $L3$, where the letters indicate the type of experiment (gas or liquid), and the numbers 1, 2 and 3 denote the corresponding Reynolds number (3,500, 6,700, and 12,000, respectively).

The trends observed in these figures generally exhibit good agreement between the model predictions and the experimental data for all the cases. In particular, our results indicate that as the magnitude of the Reynolds number and the Schmidt number increases, the decay of reactant conversion rate is slightly faster. These trends are consistent with the results of measurements by Toor (1975). In making these comparisons, however, two points should be made. First, the discrepancies at high Reynolds number may be attributed to experimental error as mentioned by Ajmera et al. (1976). Second, in all the experiments it is implied that the decay of the unmixedness is independent of the chemistry and thus is the same under both nonreacting and reacting conditions. Our results do not show this feature. To demonstrate this point, the unmixedness profile normalized by its value in the limit of no chemistry (mixing only), $[\Psi^2(Da\rightarrow\infty)]/[\Psi^2(Da=0)]$, is presented for the case $G2$ in Figure 7. Note that the asymptotic value of this normalized variable approaches the constant value of $2/\pi$. This value is due to an asymptotic Gaussian distribution for the PDF of the random variable $\mathfrak{J}$ and is consistent with that obtained in previous DNS results of Givi and McMurtry (1988) and discussed by Kosaly (1987). This limiting value is valid only if the reactants are completely segregated at the inlet of the reactor.

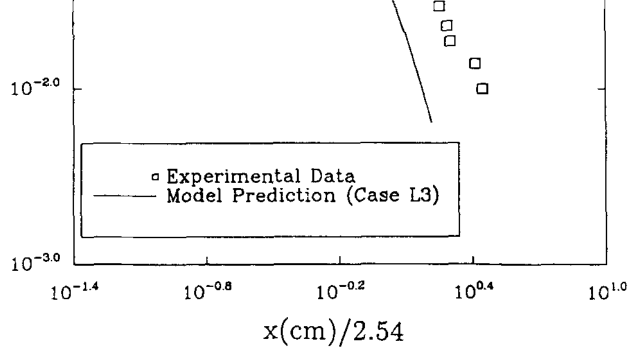

Figure 6. Reactant conversion distance in aqueous system for *Re* = 12,000.

Despite the good overall agreement between the model predictions and the experimental data, our simulations point to the need for more detailed laboratory measurements of the turbulence parameters. Since the initial turbulent integral length scale and the intensity were chosen to best reproduce the experimental data, it was deemed necessary to investigate the effects of these parameters. Therefore, a study was undertaken to determine the influences of the turbulent scale and the relative turbulent intensity on the conversion rate. For this study, the gaseous experiment with the jet Reynolds number of 6,700 was chosen.

The results of our model predictions indicate that both the integral length scale of turbulence and the turbulence intensity have a significant influence on the extent of reactant conversion. Figure 8 shows the effect of varying the turbulent integral scale (L). Three cases are considered: $L=D$, $L=D/2$, and $L=D/4$. These cases are identified by symbols $G2$, $G4$, and $G5$, respectively. The results show that generally, as the mag-

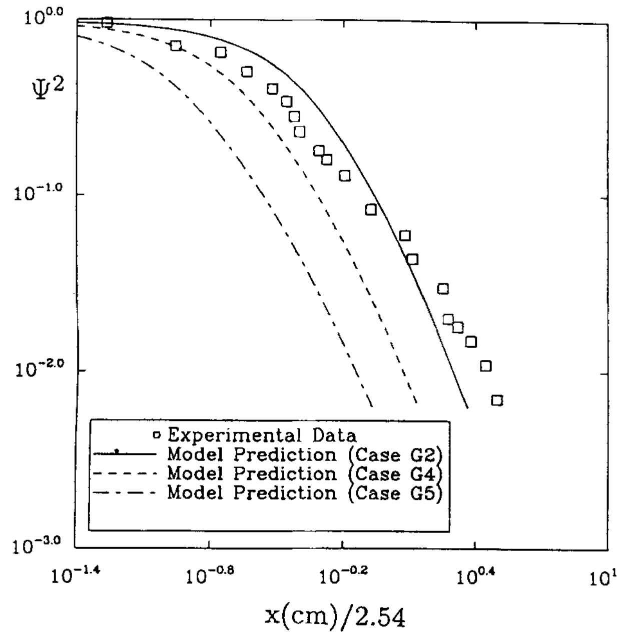

Figure 8. Reactant conversion distance in gaseous system for $Re = 6{,}700$.

nitude of the integral scale decreases, the reactant conversion occurs at a faster pace. The integral scale represents the characteristic length of the large-scale turbulent motion. For smaller L we expect smaller eddies on average and thus a faster eddy motion which in turn leads to enhanced diffusion and reaction. A somewhat analogous physical scenario is observed in the results generated by varying the magnitude of the relative turbulence intensity. This is shown in Figure 9, in which the model predictions are presented, along with the experimental data, for turbulence levels of $I = 5\%$, 10%, 15%, and 20%, identified respectively by symbols $G2$, $G6$, $G7$, and $G8$. This figure indicates that in general, the higher the level of turbulent fluctuations, the faster the decay of the unmixedness. This is also justified physically. Turbulence fluctuations serve to transport fluid blobs of one species into contact with fluid blobs of another species through eddy motion. Hence, by increasing the intensity of these fluctuations, the stirring mechanism generated by turbulence becomes more pronounced. This higher mixing results in a higher reaction, at least in an equilibrium flow, thereby resulting in a more rapid conversion rate.

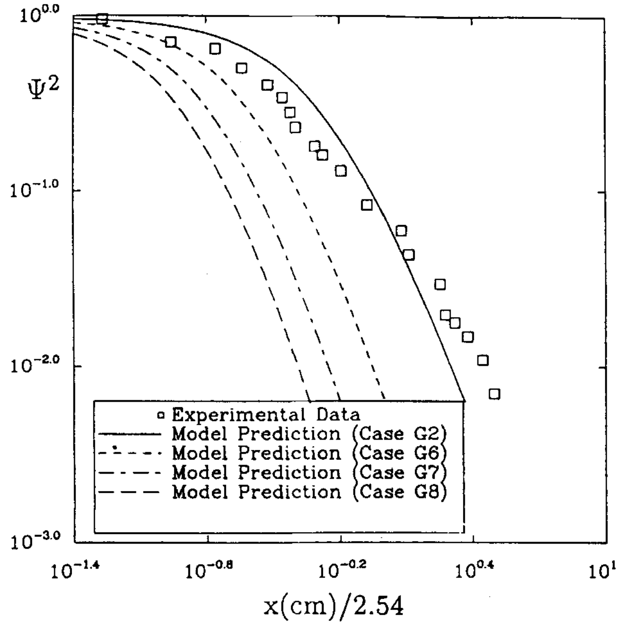

Figure 9. Reactant conversion distance in gaseous system for $Re = 6{,}700$.

The quantitative behavior presented in these figures indicates the dependence of the results on the turbulence parameters. In future experiments, it is recommended to provide a more quantitative measure of these or other relevant parameters that can be used in model validations.

Concluding Remarks

Development of the mapping closure for molecular mixing has had a significant impact on the statistical description of scalar quantities by PDF methods. In this work, we have employed this closure for predicting the limiting bound of the reactant conversion rate in a stationary isotropic turbulent flow. This has been realized for a simple chemistry of the type $A + B \rightarrow Products$ in an isothermal and constant density turbulent field. With the use of the mapping closure, the limiting bound of the conversion rate can be predicted by a simple analytic expression. This expression is very convenient due to its simplicity of utilization for modeling of plug-flow reactors (for excellent reviews on this issue, see Brodkey, 1975, 1981; Toor, 1975; and Hill, 1976). However, in the context of a single-point statistical description, the information pertaining to the evolution of the turbulent scales cannot be recovered by this expression and must be furnished by external means. To provide this information, we have implemented the EDQNM spectral closure for the physical evolution of the covariance of the relevant Shvab-Zeldovich variable characterizing the compositional structure of the reacting field.

The results predicted by the hybridized Mapping-EDQNM closure portray qualitative features similar to those of laboratory experiments and show trends in agreement with the results of previous DNS. However, in the absence of detailed experimental data, quantitative features can be matched only with laboratory data by making certain assumptions with respect to the initialization of the turbulence scales. The results of a parametric study reveal the importance of these scales for practical and realistic applications of the model.

The good overall agreement with experimental data is encouraging. This agreement is particularly noteworthy since the model employed to generate the data is rather simple and is very easy to implement computationally. Most of the computational efforts are those associated with the numerical solution of the EDQNM closure. In a laboratory experiment, these calculations can simply be replaced with the measurements of the decay of the covariance of a conserved scalar. With such measurements, our closed form algebraic relation can then be used for an efficient and reasonably effective estimate of the limiting bound of the conversion rate.

There are many ways by which the model can be improved. It would be desired to extend the hybrid methodology for less restrictive and more physically complex reacting flow systems;

for example, nonequilibrium, exothermic chemistry in variable density, decaying turbulent fields. In such circumstances, the extension of the mapping closure is straightforward since this closure is based on the PDF of the scalar quantities and can account for the effects of complex chemistry. However, the results may not be presentable by simple analytical relations as derived here and would require numerical solution of the mapping function (Valiño and Gao, 1991). Also, the development of EDQNM for compressible turbulence is somewhat more involved, but has been recently initiated (Marion et al., 1989). These new developments can be utilized in capturing the effects of compressibility in reacting turbulence phenomenon.

In future application, the assumption of spatial isotropy may be relaxed. The EDQNM model can be used in homogeneous (but nonisotropic) flows. Also it is possible to implement the model for a decaying turbulence field. In fact, this model was originally developed for the hydrodynamic closure before its application for transport of scalar quantities (Lesieur, 1991). Therefore, the assumption of stationary turbulence imposed in these simulations can be relaxed. In the present work, this was not deemed necessary due to the accuracy range of the experimental data. Finally, the extension of the mapping closure to account directly for the length scale information can be accomplished by an appropriate mapping procedure for the joint PDF of the scalar(s) and its (their) gradient(s). However, such a mapping is very complicated and still requires certain external information for a complete closure (Chen et al., 1989; Pope, 1991). Some issues regarding the shortcomings of the single-point mapping closure in predicting the statistics of the gradient field are discussed by Gao (1991b) and Jiang et al. (1992).

In comparison with currently available alternatives, our model seems very attractive, even with the imposition of several restricting assumptions. This is primarily due to a firm mathematical-physical basis of the model and the simplicity of the final results. As such, the procedure is plausible in both basic research on the physics of reacting turbulent transport and practical applications in the modeling and design of plug-flow reactors. Current work by Kraichnan on further extension of the mapping closure and identification of its capabilities and drawbacks would be very useful in the future work in this direction.

Acknowledgment

This work is supported by the National Science Foundation under Grant CTS-9012832, and by the Office of Naval Research under Grant N00014-90-J-4013. Acknowledgment is also made to the Donors of the Petroleum Research Funds administrated by the American Chemical Society for partial support of the authors under Grant ACS-PRF#25129-AC7E,6. Computational support is provided by NCSA at the University of Illinois.

Notation

A,B = reactants concentration
a,b = fluctuating components of reactant concentrations
C_1,C_2,C_3 = constants
D = reactor tube diameter
$\mathcal{D}$ = molecular diffusivity of species
Da = Damköhler number
E = turbulent energy spectrum function
H = Heaviside function
I = relative turbulence intensity
$\mathcal{J}$ = Shvab-Zeldovich conserved scalar variable
$J_{3/2}$ = Bessel function of 3/2 order
$\mathcal{K}$ = turbulent eddy diffusivity
L = hydrodynamic integral scale
$L_{\mathcal{J}}$ = scalar integral scale
$\mathcal{P}$ = PDF
Re = Reynolds number
r = physical radial coordinate
Sc = molecular Schmidt number
t = physical time
x = spatial coordinate (downstream distance from the reactor inlet)

Greek letters

γ = constant in EDQNM closure
δ = delta function
η = composition space for a Gaussian reference field
Θ = triad interaction time
κ = wave number in spectral domain
λ = Taylor microscale for the hydrodynamic field
$\lambda_{\mathcal{J}}$ = Taylor microscale for the Shvab-Zeldovich variable
λ'' = constant in EDQNM closure
μ'' = eddy relaxation frequency
ν = kinematic viscosity
ρ = scalar covariance
Σ = parameter in mapping closure
χ = mapping function
Ψ^2 = unmixedness parameter
ψ = composition domain

Subscripts

0 = time zero (inlet of plug-flow reactor)
G = Gaussian

Other symbols

$\langle\ \rangle$ = ensemble average
$\tilde{}$ = normalized value

Literature Cited

Ajmera, P. V., M. Singh, and H. L. Toor, "Reactive Mixing in Turbulent Gases," *Chem. Eng. Commun.*, **2**, 115 (1976).

Bilger, R. W., "Turbulent Flows with Nonpremixed Reactants," *Turbulent Reacting Flows*, P. A. Libby and F. A. Williams, eds., Chap. 3, Vol. 44, Topics in Applied Physics, Springer-Verlag Berlin, Heidelberg (1980).

Brodkey, R. S., ed., "Mixing in Turbulent Fields," *Turbulence in Mixing Operations*, Academic Press, New York (1975).

Brodkey, R. S., "Fundamentals of Turbulent Motion, Mixing and Kinetics," *Chem. Eng. Commun.*, **8**, 1 (1981).

Chen, H., S. Chen, and R. H. Kraichnan, "Probability Distribution of a Stochastically Advected Scalar Field," *Phys. Rev. Lett.*, **63**, 2657 (1989).

Engels, H., *Numerical Quadrature and Cubature*, Academic Press, New York (1980).

Eswaran, V., and E. E. O'Brien, "Simulations of Scalar Mixing in Grid Turbulence Using an Eddy-Damped Closure Model," *Phys. Fluids A*, **1**(3), 537 (1989).

Eswaran, V., and S. B. Pope, "Direct Numerical Simulations of the Turbulent Mixing of a Passive Scalar," *Phys. Fluids A*, **31**, 506 (1988).

Gao, F., "Theoretical and Numerical Investigations of Scalar Fields in Isotropic Turbulence," PhD Thesis, State University of New York at Stony Brook (1990).

Gao, F., "An Analytical Solution for the Scalar Probability Density Function in Homogeneous Turbulence," *Phys. Fluids A*, **3**(4), 511 (1991a).

Gao, F., "Mapping Closure and Non-Gaussianity of the Scalar Probability Density Functions in Isotropic Turbulence," *Phys. Fluids A*, 3(10) (1991b).

Givi, P., "Model Free Simulations of Turbulent Reactive Flows," *Prog. Energy Combust. Sci.*, **15**(1), 1 (1989).

Givi, P., and McMurtry, P. A., "Nonpremixed Reaction in Homogeneous Turbulence: Direct Numerical Simulations," *AIChE J.*, **34**(6), 1039 (1988).

Hawthorne, W. R., D. S. Wedell, and H. C. Hottel, "Mixing and Combustion in Turbulent Gas Jets," *Symp. on Combustion, Flames and Explosion Phenomena*, p. 266, Combustion Inst., Pittsburgh (1949).

Herring, J. R., D. Schertzer, M. Lesieur, G. R. Newman, J. P. Chollet, and M. Larcheveque, "A Comparative Assessment of Spectral Closures as Applied to Passive Scalar Diffusion," *J. Fluid Mech.*, **124**, 411 (1982).

Hill, J. C., "Homogeneous Turbulent Mixing with Chemical Reaction," *Ann. Rev. Fluid Mech.*, **8**, 135 (1976).

Janicka J., W. Kolbe, and W. Kollmann, "Closure of the Transport Equation for the Probability Density Function of Turbulent Scalar Fields," *J. Non-Equilib. Thermodyn.*, **4**, 47 (1979).

Jiang, T.-L., "Turbulent Mixing in Nonreacting and Reacting Flows: a PDF Approach at the Two-Point Level," PhD Thesis, State University of New York at Stony Brook (1990).

Jiang, T.-L., F. Gao, and P. Givi, "Binary and Trinary Scalar Mixing by Fickian Diffusion—Some Mapping Closure Results," *Phys. Fluids A.*, in press (1992).

Kollmann, W., "The PDF Approach to Turbulent Flow," *Theoret. Comput. Fluid Dynamics*, **1**, 249 (1990).

Kosaly, G., "Nonpremixed Simple Reaction in Homogeneous Turbulence," *AIChE J.*, **33**(12), 1998 (1987).

Kosaly, G., and P. Givi, "Modeling of Turbulent Molecular Mixing," *Combust. Flame*, **70**, 101 (1987).

Larcheveque, M., and M. Lesieur, "The Application of Eddy-Damped Markovian Closures to the Problem of Dispersion of Particle Pairs," *J. Mech.*, **20**, 113 (1981).

Launder, B. E., and D. B. Spalding, *Lectures in Mathematical Models of Turbulence*, Academic Press, New York (1972).

Lesieur, M., *Turbulence in Fluids*, Nijhoff, Dordrecht (1990).

McMurtry, P. A., and P. Givi, "Direct Numerical Simulations of Mixing and Reaction in a Nonpremixed Homogeneous Turbulent Flow," *Combust. Flame*, **77**, 171 (1989).

Madnia, C. K., S. H. Frankel, and P. Givi, "Mathematical Modeling of the Reactant Conversion Rate by Single-Point PDF Methods," *Proc. of Combustion Inst.*, Ithaca, NY (1991).

Marion, J. D., J. P. Bertoglio, and J. Mathieu, "An Eddy Damped Quasi Normal Markovian Approach to Compressible Isotropic and Homogeneous Turbulence," *Advances in Turbulence 2*, H. H. Fernholz and H. E. Fielder, eds., Springer-Verlag Berlin, Heidelberg (1989).

Millionshtchikov, M., "On the Theory of Homogeneous Isotropic Turbulence," *C. R. Acad. Sci. U.R.S.S.*, **32**, 615 (1941).

Miyawaki, O., H. Tsujikawa, and Y. Uraguchi, "Turbulent Mixing in Multi-Nozzle Injector Tubular Mixer," *J. Chem. Eng. Japan*, **7**, 52 (1974).

Norris, A. T., and S. B. Pope, "Turbulent Mixing Model Based on Ordered Pairing," *Combust. Flame*, **27**, 83 (1991).

O'Brien, E. E., "Turbulent Reacting Flows," *Topics in Applied Physics*, P. A. Libby and F. A. Williams, eds., Chap. 5, Vol. 44, Springer-Verlag Berlin, Heidelberg (1980).

Orszag, S. A., "Statistical Theory of Turbulence," *Fluid Dynamics*, R. Balian and J. L. Peabe, eds., Gordon and Breach, New York (1977).

Pope, S. B., "The Statistical Theory of Turbulent Flames," *Phil. Trans. R. Soc. Lond.*, **291**, 529 (1979).

Pope, S. B., "PDF Methods for Turbulent Reacting Flows," *Prog. Energy Combust. Sci.*, **11**, 119 (1985).

Pope, S. B, "An Improved Turbulent Mixing Model," *Combust. Sci. Tech.*, **28**, 131 (1982).

Pope, S. B., "Computations of Turbulent Combustion: Progress and Challenges," *Proc. of Int. Symp. on Combustion*, The Combustion Institute, Pittsburgh, p. 591 (1990).

Pope, S. B., "Mapping Closures for Turbulent Mixing and Reaction," *Theoret. Comput. Fluid Dynamics*, **2**, 255 (1991).

Tavoularis, S., and S. Corrsin, "Experiments in Nearly Homogeneous Turbulent Shear Flow With a Uniform Mean Temperature Gradient," *J. Fluid Mech.*, **104**, 311 (1981).

Tennekes, H., and J. L. Lumley, *A First Course in Turbulence*, MIT Press, Cambridge, MA (1972).

Toor, H. L., "Mass Transfer in Dilute Turbulent and Nonturbulent Systems with Rapid Irreversible Reactions and Equal Diffusivities," *AIChE J.*, **8**(1), 70 (1962).

Toor, H. L., "The Nonpremixed Reaction: $A + B \rightarrow$ *Products*," *Turbulence in Mixing Operations*, R. S. Brodkey, ed., Academic Press, New York (1975).

Valiño, L., and F. Gao, "Monte Carlo Implementation of the Mapping Closure for Turbulent Reacting Flows," Amer. Phys. Soc., Phoenix, AZ (Nov. 1991).

Manuscript received Aug. 26, 1991, and revision received Feb. 7, 1992.