Nonpremixed Reaction in Homogeneous Turbulence: Direct Numerical Simulations

Peyman Givi
Department of Mechanical and
Aerospace Engineering
State University of New York
Buffalo, NY 14260

Patrick A. McMurtry
Division 8363
Sandia National Laboratories
Livermore, CA 94550

In accordance with Toor's original paper (1962), consider an irreversible, one-step isothermal reaction of the type $A + B \rightarrow$ Products in a nonpremixed homogeneous turbulent flow. Let $A(\underline{x}, t)$ and $B(\underline{x}, t)$ define the instantaneous concentration of the two species and decompose each concentration into an ensemble mean value, denoted by $\langle \ \rangle$, and its fluctuation from the mean value, represented by lowercase letters:

$$\begin{aligned} A &= \langle A \rangle + a \\ B &= \langle B \rangle + b \end{aligned} \tag{1}$$

Under the assumptions given above, the transport equations governing the evolution of the mean concentrations are

$$L(\langle A \rangle) = \frac{d\langle A \rangle}{dt} = -W \tag{2}$$

$$L(\langle B \rangle) = -W \tag{3}$$

$$W = K[\langle A \rangle \langle B \rangle + \langle ab \rangle] \tag{4}$$

where W is the mean reaction rate, K is the kinetic speed of the reaction, and L is the convection-diffusion operator, which is the same for the two reactants if their diffusion coefficients are identical. The second term on the righthand side of Eq. 4 is unclosed and needs to be modeled. This term can be written, in normalized form (Toor, 1975; Kosaly, 1987),

$$\Psi^2(t) = \frac{\langle ab \rangle_t}{\langle ab \rangle_o} \tag{5}$$

where the subscripts t and o refer to the time and the initial time (i.e., the inlet of a plug-flow reactor), respectively. Turbulent mixing is characterized by the root mean square of one of the reactants in the absence of reaction:

$$d^2(t) = \frac{\langle a_m^2 \rangle_t}{\langle a_m^2 \rangle_o} \tag{6}$$

where m refers to "pure mixing" only. It is obvious that, in the limit of zero rate chemistry, $d^2(t)$ and $\Psi^2(t)$ are identical (Toor, 1975), i.e.,

$$d^2(t) = \lim_{K \to 0} \Psi^2(t) \tag{7}$$

In the limit of infinitely fast chemistry under stoichiometric conditions, and assuming a Gaussian shape for the probability density function (PDF) of the mixture fraction $J(\underline{x}, t)$ [defined by $J(\underline{x}, t) = A(\underline{x}, t) - B(\underline{x}, t)$], Toor showed that

$$d^2(t) = \lim_{K \to \infty} \Psi^2(t) \tag{8}$$

(The assumption of Gaussian shape can be relaxed for symmetric PDF's [O'Brien, 1971].)

Based on Eqs. 7 and 8, Toor assumed that

$$\Psi^2(t) = d^2(t) \text{ for any } K \tag{9}$$

Equation 9 summarizes Toor's hypothesis.

In the recent paper of Kosaly (1987), it is shown that the application of Toor's hypothesis is not valid for the predictions of plug-flow reactors in which the initial shape of the PDF of the

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Correspondence concerning this paper should be addressed to Peyman Givi.

variable $J(\underline{x}, t)$ is not Gaussian but rather of bimodal shape determined by initial conditions. Kosaly showed that, while $d^2(t)$ and $\Psi^2(t)$ are identical for zero rate chemistry (i.e., Eq. 7) and at $t = 0$ for all K, Eq. 8 should be replaced by

$$\lim_{K\to\infty} \Psi^2(t) = \frac{2}{\pi} d^2(t) \tag{10}$$

at later times. The factor $2/\pi$ was a result of algebraic manipulation assuming the PDF of J becomes Gaussian; see Kosaly (1987) for details.

In this note, we first provide a generalization of Kosaly's result and, by use of direct numerical simulation (DNS), further investigate the validity of Toor's hypothesis and the revisions proposed by Kosaly. In this application DNS refers to the numerical solution of the aerothermodynamic transport equations of turbulent reacting flows for the detailed time development of the flow field. This technique uses no closure modeling, and no assumptions are made pertaining to the turbulent behavior of the fluid. DNS has proven very useful in computational studies of turbulence (see Rogallo and Moin, 1984, for a review) and is used here as a means of performing experiments on the computer. This approach offers advantages over laboratory experiments in that the instantaneous values of the hydrochemical variables are known at all locations at each time step, allowing better statistical analysis of the data. Computer limitations of course restrict the ranges of time and space scales that can be accurately resolved.

Generalization of Toor's Hypothesis

Kosaly's results can be generalized by defining the normalized variable $\hat{\phi}$ and the standard PDF $\hat{P}(\hat{\phi}, \hat{t})$ as (Pope, 1982)

$$\hat{\phi} = \frac{\phi}{\sigma(t)} \tag{11}$$

$$\hat{P}(\hat{\phi}, t) = P(\phi, t)\sigma(t) \tag{12}$$

where $P(\phi, t)$ is the PDF of J, and σ is the standard deviation. Assuming that the PDF approaches as asymptotic time-independent form (whether Gaussian or not), we have

$$\hat{P}(\hat{\phi}, t) \approx \hat{P}(\hat{\phi}) \tag{13}$$

or

$$P(\phi, t) \approx \frac{\hat{P}[\phi/\sigma(t)]}{\sigma(t)} \tag{14}$$

With an asymptotic shape of $\hat{P}(\hat{\phi})$, Kosaly's results generalize to

$$\Psi^2(t) = Cd^2(t) \tag{15}$$

where

$$C = 4\left[\int_0^\infty \hat{\phi}\hat{P}(\hat{\phi})\, d\hat{\phi}\right]^2 \tag{16}$$

For a PDF that asymptotically assumes a Gaussian form,

$$\hat{P}(\hat{\phi}) = \frac{1}{\sqrt{2\pi}} \exp(-\hat{\phi}^2/2) \tag{17}$$

the value of C would be the same as that given by Kosaly (1987), i.e., $C = 2/\pi$. The exact asymptotic shape of the PDF therefore determines the constant of proportionality in Eq. 15. The magnitude of this constant, which is valid only in the final stages of mixing, cannot be predicted mathematically and can only be estimated experimentally or numerically.

Simulation Results

A pseudospectral numerical method developed by McMurtry (1987) was modified for the calculations of the homogeneous turbulent flow considered here. This method is very similar to that employed previously by Riley et al. (1986), McMurtry et al. (1986, 1987), Givi et al. (1987), and Givi and Jou (1987), and therefore will not be explained here. The scalar fields are defined to be square waves with the two reactants out of phase and at stoichiometric conditions. This is similar to that used previously be Leonard and Hill (1986, 1987). The flow field is initialized by specifying the turbulence spectrum (similar to that used by McMurtry, 1987), and a forcing mechanism is employed to keep the turbulence stationary by adding energy artificially to the large-scale motions (low wave numbers in Fourier space). This forcing was applied in such a way as to compensate for small-scale dissipation without affecting the small-scale statistics of interest (McMurtry and Givi, 1987). Periodic boundary conditions are employed in all three directions, and the aerothermodynamical variables are spectrally approximated on 64 × 64 × 64 collocation points within the computational domain. The values of the Reynolds and Peclet numbers are kept within moderate levels to ensure the accuracy of the simulations on the grids used. Calculations were performed with zero rate and infinitely fast chemical reactions to assess the influence of the chemistry on the decay rate of the unmixedness parameter Ψ^2. In the zero rate chemistry simulations, the value of K was set to zero in Eq. 4, whereas in the infinitely fast rate chemistry calculations the statistical variations of the two reactants A and B were related to that of the conserved scalar variable J (Bilger, 1980; Kosaly and Givi, 1987).

For the purpose of flow visualization, two-dimensional contour plots of species A are presented in Figure 1. Parts (*a*) and (*b*) of this figure correspond to zero rate and infinitely fast chemical reactions, respectively. Time is normalized by the large-scale turbulence frequency, and the nondimensional time of $t = 1.2$ corresponds to the case when the average concentration of species A under reacting conditions has reduced to 30% of its initial value. This figure exhibits the effects of three-dimensional turbulent motion on the distortion of the scalar field and the mixing of the two initially segregated reactants (the contours form parallel lines at $t = 0$). The effects of chemical reactions are to increase the steepness of the scalar gradients and, obviously, to reduce the instantaneous values of the reactants as indicated by a comparison of Figures 1a and 1b. The influence of the chemical reactions on the decay rate of the unmixedness is shown in Figure 2. In this figure, the ratio $\Psi^2(t)/d^2(t)$ is presented vs. time for zero rate and infinitely fast chemical reactions. For zero rate chemistry, the ratio obviously

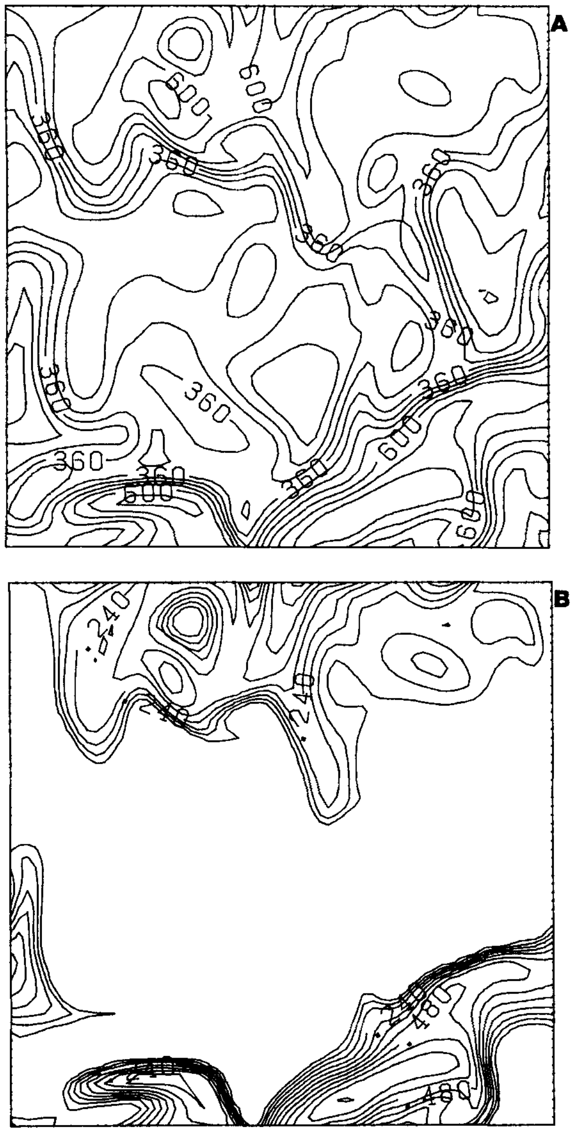

Figure 1. Plots of species A concentration contours at $t = 1.2$.

(a) Nonreacting case. Contour minimum, 0.06; contour maximum, 0.72; contour interval, 0.06; labels scaled by 1,000.

(b) Reacting case. Contour minimum, 0; contour maximum, 0.66; contour interval, 0.06

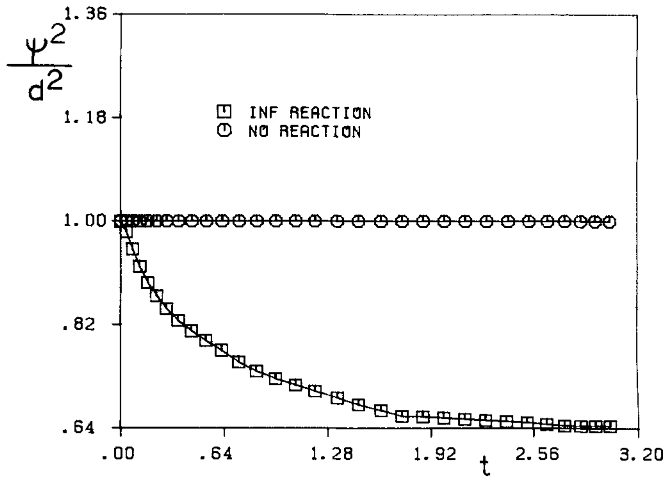

Figure 2. Temporal variation of Ψ^2/d^2.

remains at unity. For the reacting case, at the initial time ($t = 0$), the influence of chemistry is nil. At later times, however, the value of the unmixedness for the fast chemistry is lower than that under zero rate chemistry. The results shown in Figure 2 indicate that for initially nonpremixed reactants, the decay rate of the unmixedness is not independent of the chemical reactions and depends on the magnitude of the local Damkohler number.

The theoretical discussion given in the previous section on the lack of agreement with Toor's hypothesis is verified by examining the temporal evolution of the PDF of J (i.e., $P(\phi, t)$, $-1 \leq \phi \leq 1$) in Figure 3. It is clearly seen that the evolution of the PDF from its initial bimodal shape (composed of two delta functions at $\phi = \pm 1$ to its asymptotic shape (composed of a single delta function at $\phi = 0$) cannot be characterized by its first two moments (i.e., 0 and σ). Therefore, Eq. 9 is not valid for intermediate times, and the application of Toor's hypothesis is not appropriate for the prediction of the conversion rate in plug-flow type reactors with initially segregated reactants.

Figure 2 further indicates the interesting result that the ratio of Ψ^2/d^2 for the infinitely fast chemistry approaches the value of 0.64 ($\approx 2/\pi$). This value corresponds to the case if $\hat{P}(\phi)$ asymptotically adopts a Gaussian profile (Kosaly, 1987). To ascertain the asymptotic shape of the PDF, the temporal variation of the kurtosis (μ_4 = normalized fourth moment) of species A is presented in Figure 4. It is seen that, under no chemical reaction, the value of this variable approaches $\mu_4 = 2.9$. The asymptotic values of the normalized sixth (superskewness) and eighth moments are $\mu_6 = 13.9$ and $\mu_8 = 98$, and all the odd moments remain close to zero (not shown here). A comparison of these values with those of a Gaussian profile (i.e., $\mu_4 = 3$, $\mu_6 = 15$, and $\mu_8 = 105$) indicates that the approximation of a Gaussian PDF for the final stages of mixing of a nonreacting scalar is justified. Better agreements would require a larger sample size than the presently used 64^3 data points for more accurate statistical analysis. For the reacting case, however, the values of the normalized fourth- and higher order moments are larger than those of

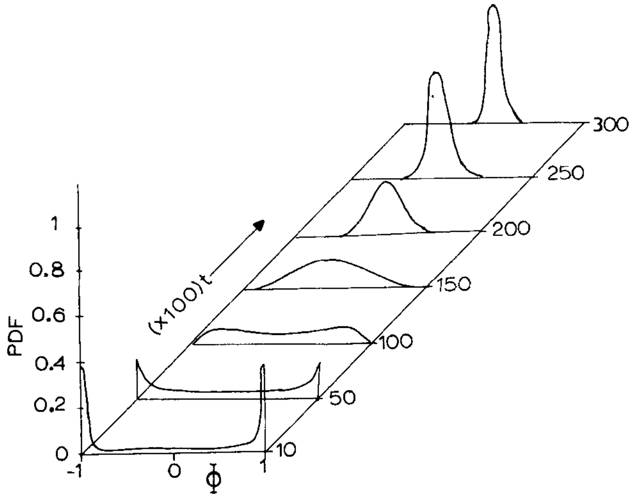

Figure 3. Temporal variation of $P(\phi)$.

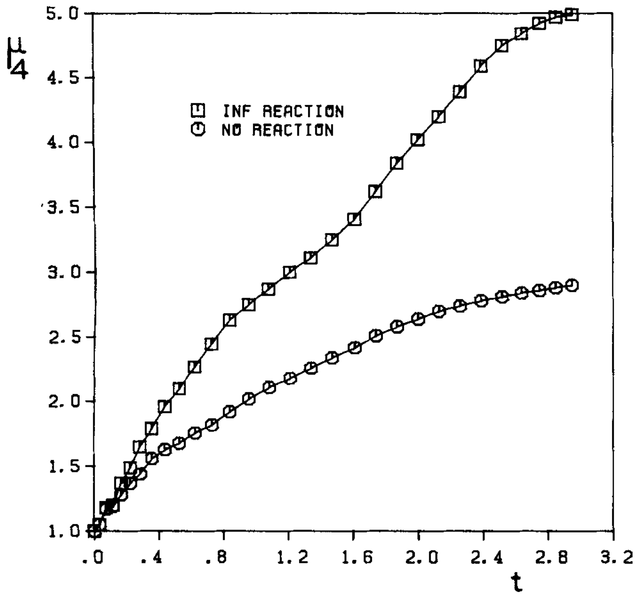

Figure 4. Temporal variation of μ_4.

an inert scalar. This is shown in Figure 4 for the kurtosis of one of the reactants and is consistent with our previous calculations (Kosaly and Givi, 1987). Therefore, a Gaussian PDF assumption is not justified in this case.

Finally, it must be mentioned that the results of our present calculations are not in agreement with those previously obtained by Leonard and Hill (1986). In their calculations, the ratio of Ψ^2/d^2 initially decreases from unity with time buy asymptotically returns to unity for longer times. This lack of agreement with Leonard and Hill's calculations may be attributed to the fact that in their finite rate chemistry simulations, the magnitude of the Damkohler number is not large enough to justify the infinitely fast chemistry assumption adopted here.

The results of this work and those of Kosaly (1987) indicate the need for detailed measurements in plug-flow reactors for comparison with numerical simulations. The present simulations indicate that Toor's hypothesis ($\Psi^2 = d^2$) should be modified by Eq. 15 for infinite rate chemistry at the temporal asymptote. For finite rate chemistry and intermediate times, however, the application of the hypothesis is not valid and full simulations are required. The numerical experiments reported here indicate that the asymptotic shape of the PDF of a conserved scalar characterizing mixing is approximately Gaussian. This suggests a value of $C = 2/\pi$ in Eq. 15.

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Notation

A, B = instantaneous concentration of reacting species
a, b = instantaneous concentration fluctuation of reacting species
$d^2(t) = \langle a_m^2 \rangle_t / \langle a_m^2 \rangle_o$
$J = A - B$
K = kinetic reaction rate
P = PDF
t = time
W = reaction rate
σ = standard deviation
ϕ = scalar space
$\Psi^2(t) = \langle ab \rangle_t / \langle ab \rangle_o$

Indices and symbols

o = initial
m = mixing in absence of chemical reaction
$\langle\ \rangle$ = ensemble average

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