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PDF CALCULATIONS OF MAJOR AND MINOR SPECIES IN A TURBULENT PILOTED JET FLAME

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The joint probability density function (JPDF) approach is used to make calculations of a piloted jet diffusion flame of CH_4-air (flame L of Masri et al. [1]). The calculations include detailed chemistry, involving 16 species and 41 reactions, to account for the finite-rate kinetics. The technique of *in situ* adaptive tabulation (ISAT [2,3]) enables the computationally efficient inclusion of detailed chemistry in the JPDF calculations.

The results of these JPDF-ISAT calculations show good agreement with the experimental data, including those for the mean and rms of minor species such as CO and H_2. These results are discussed in the context with the earlier numerical studies of this flame ([4–6]). The present calculations of minor species are significantly better than those from earlier studies.

Introduction

Joint probability density function (JPDF) methods for turbulent-reactive flows [7] have the natural advantage of providing an exact representation of the nonlinear reaction and turbulent convection. The Monte Carlo method has been successfully applied to solve the velocity-frequency-composition joint PDF equations [8] and has been implemented for various flows (e.g., [5,9,10]). JPDF calculations of piloted turbulent non-premixed flames have been attempted in the past by Masri and Pope [5], Masri et al. [11] and Norris and Pope [10] to demonstrate the application of the velocity-frequency-composition JPDF method. The conserved scalar approach [12] was used in the first work while an assumption of self-similar chemistry was made for the second study. The third study used the intrinsic low-dimensional manifold (ILDM) technique [13] for reducing chemistry. Hybrid methods involving composition PDF with a CFD (computational fluid dynamics) method for velocity have been used for such flames as well (e.g., [4,6,14]).

The common feature of the foregoing studies is that the chemistry was included in these calculations at a relatively simple level because the CPU requirements for JPDF calculations with detailed chemistry were deemed to be prohibitively expensive.

However, the computational task of including detailed chemistry in JPDF calculations can now be accomplished efficiently using the *in situ* adaptive tabulation (ISAT) algorithm developed by Pope [2]. ISAT has been shown to reduce the computer time needed to solve the reaction equations by up to 3 orders of magnitude with reasonable storage requirement and with good control of interpolation errors [2,15]. The performance of ISAT in JPDF calculations has been detailed in a recent study [3].

In the next two sections, the main features of the flame selected for this study and of the previous calculations of this flame are presented. This is followed by the numerical details of our calculations and comparison of our results with the experimental data as well as with other calculations. We conclude by evaluating our calculations in view of these comparisons.

The Flame Considered

We consider flame L of Masri et al. [1], which is a piloted jet diffusion flame of methane air, for which sufficient data for velocity and thermochemistry are available. It is reported to be far from extinction and is relatively free from soot up to about 30 jet diameters downstream of the nozzle exit.

The radius of the central jet of the piloted burner is 3.6 mm, and that of the outer annulus of the pilot is 9 mm. The fuel is pure methane, and the outer coflow is air. The fuel and air are at 300 K and have velocities of 41 and 15 m/s, respectively. A peculiar feature of the flame is that the pilot is a stoichiometric, premixed, and fully burned annular flow of C_2H_2, H_2 and air with the C/H ratio adjusted to be the same as that of the fuel. Consequently, there is excess enthalpy in the pilot compared to a fully burned stoichiometric mixture of methane and air.

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The initial conditions for mean and fluctuating velocities are well documented [1]. The flame was investigated using both probe and Raman–Rayleigh measurements. No species-specific LIF (laser-induced fluorescence) was used.

Previous Calculations

The main features of the previous calculations of flame L are briefly described here. Chen et al. [4] have used a hybrid scheme that employs a composition PDF method for scalars with a second-moment closure method for modeling turbulent velocity statistics. Two reduced mechanisms involving four and five scalars were used for modeling reaction and a modified coalescence-dispersion model was used for modeling mixing. Chen et al. found that to avoid the blowout of the flame, they had to "artificially ignite" the flame by inserting particles with burned stoichiometric mixture into the fuel jet just downstream of the jet pipe exit. The radial profiles for volume-averaged density, temperature, and water were presented. The scatter plots of CO suggested substantial underprediction with both chemistry models.

Masri and Pope [5] solved for the JPDF of velocity-frequency composition using the IEM (interaction by exchange with the mean) mixing model and with two variants of simple thermochemistry: (a) density is assumed a piecewise function of mixture fraction and (b) flamelet chemistry. Radial profiles for only two major species were plotted in addition to density, mixture fraction, and temperature. The need to include a more detailed description of chemistry was clearly recognized in this study.

A hybrid CFD–Eulerian PDF scheme is used by Zurbach et al. [6] with modified coalescence-dispersion mixing model and a three-dimensional ILDM table for chemistry. The $\kappa-\varepsilon$ model is used for turbulence in the CFD code. Radial profiles for major and minor species are presented by the authors. Major species are generally well predicted, but minor species do not show good agreement. Zurbach et al. report that with the IEM mixing model, the flame blew out.

The Favre-averaged results for species concentration from the latter two studies, where available, are compared later with our JPDF calculations and the experimental data.

Description of the JPDF Model

A modeled transport equation is solved for the joint PDF of velocity, turbulent frequency, and thermochemical composition. All of the submodels used are standard and have been described in previous works. The simplified Langevin model (SLM [7]) is used for velocity, with the standard value of the model constant $C_0 = 2.1$. The turbulence frequency model used is that of Jayesh and Pope [16]. Again, standard values of the constants are used, namely, $C_1 = 0.08$, $C_2 = 0.9$, $C_3 = 1.0$, and $C_4 = 0.25$.

Initially, calculations were performed using the IEM model but, consistent with previous studies (e.g., Refs. [11] and [6]), it was found that the flame did not stay lit. The EMST (Euclidian minimum spanning tree) based model [17] seems better to incorporate the physics of mixing in non-premixed flames. With the EMST model, no artificial ignition was found to be necessary. The following results are obtained using the EMST model with the standard constant $C_\phi = 2.0$.

The reaction mechanism used is the skeletal C_1 mechanism [18] for methane reported in Ref. [19], which involves 4 elements, 16 species, and 41 reactions. While the use of ISAT allows the incorporation of much more detailed chemistry than reduced mechanisms, it should nevertheless be acknowledged that a C_2 mechanism is required to represent realistically the combustion of rich mixtures. In view of element conservation (and the assumption of equal species molecular diffusivities), only 13 of the species are linearly independent in the skeletal mechanism. Because the pressure is essentially constant (i.e., atmospheric pressure), the thermochemical composition can be described by 14 variables—13 species mass fractions and the enthalpy. Because the enthalpy is represented explicitly, the enthalpy excess of the pilot stream can be accounted for without difficulty.

Numerical Method

The modeled joint PDF equation is solved by the Lagrangian particle/mesh method incorporated in the code PDF2DV [8]. The particle evolution equations are integrated in time (over a time step Δt) until the statistically stationary state is reached. In each time step, separate fractional steps are used to move the particles with their own velocity, advance the particle velocity according to the simplified Langevin model, advance the turbulent frequency, perform mixing according to the EMST model, and to advance the thermochemical composition according to the kinetic mechanism. The accuracy of the splitting of mixing and reaction has been verified by Yang and Pope [19]. Details of the computations are given in Table 1.

The fractional step corresponding to reaction is performed using the ISAT algorithm [2]. The ISAT tolerance was chosen from the detailed error analysis [3] to calculate major species within 2% and minor species within 10% of the values obtained by exactly integrating the reaction equations.

Over the entire calculation, the reaction fractional step is performed over 50 million times (i.e., for each

TABLE 1
Parameters used in the JPDF calculations. R is the fuel jet radius and n_x and n_y are the number of grid nodes in axial and radial directions, respectively.

Domain size	X/R, Y/R	60.0, 15.0
Grid discretization	$n_x \times n_y$	31×31
Max. particles per cell	N_{pc}	100
Time step	Δt	0.02 ms
Number of time steps	N	700
Fuel jet bulk speed	U_j	41 m/s
Pilot speed	U_p	24 m/s
Coflow air speed	U_c	15 m/s
ISAT error tolerance	ε_{tol}	0.0008

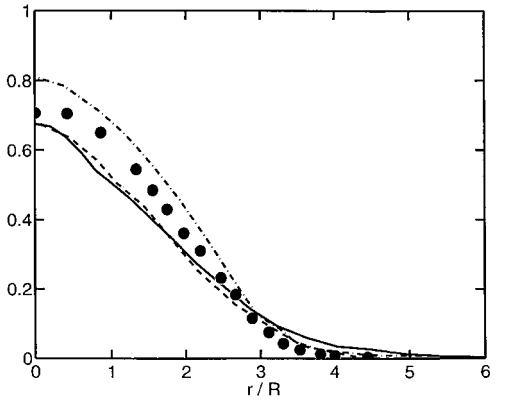

FIG. 1. Radial distribution of the Favre-mean mixture fraction at $x/R = 40$, where R is the radius of the fuel pipe. Symbols represent data from Masri et al. [1]. Continuous line is for the present JPDF–ISAT calculations. Dashes denote the results of Zurbach et al. [6], and dash-dots are for Masri and Pope [5].

of about 72,000–100,000 particles on each of about 700 time steps). Compared to integrating the reaction equations numerically, the use of the ISAT algorithm yielded a speedup of a factor of 35.

Overall, the CPU time taken by the chemistry computations alone was reduced to about 60% of the total CPU time. The ISAT table size was about 50 Mbytes, and the CPU time for the entire simulation on an SGI Indigo-2 workstation was about 25 h.

The simulation is allowed to run for long enough so that the flow achieves a statistically stationary state. The evolution of different flow quantities and mole fractions of major and minor species are monitored at different locations in the domain to determine when statistical stationarity is achieved. This occurs after about 700 time steps. The nodal values of fields (e.g., temperature, compositions, etc.) for every fourth time step, for the next 100 time steps, were stored to perform time averaging to reduce the statistical fluctuations in the profiles subsequently presented.

After these calculations were performed, Xu and Pope [20] conducted a comprehensive study of numerical errors in the JPDF algorithm applied to this flame. An unexplained finding of this study is that there is a partial cancellation of the deterministic errors—truncation error and bias. Based on that work [20], we estimate that if the same cancellation of error occurs, the numerical errors in the present calculation of the mean velocity and mixture fraction are only a few percent. However, if this cancellation of error does not occur, our estimate of numerical errors for mean velocity is about 8% and for mean mixture fraction is about 15%. Furthermore, Subramaniam [21] found (for a substantially different test case) that the number of particles per cell needed to give numerically accurate calculations for the EMST mixing model is greater than that used here. Further work is in progress to quantify these numerical errors.

Results and Discussion

The radial profiles of the Favre-averaged quantities at $x/R = 40$ are plotted in Figures 1, 2, 4, 5, and 6. The continuous lines in these figures correspond to the present JPDF-ISAT calculations. The dashed lines represent the recent results from Zurbach et al. [6], and the dash-dots are for those of Masri and Pope [5]. Symbols denote the experimental data. All quantities are Favre averaged. The results of Chen et al. [4] are not included in the plots because they are for volume-averaged quantities.

Mixture Fraction

Figure 1 shows the mean mixture fraction profiles from three different calculations compared to the experimental data. In the central region of the flame, Masri and Pope's calculations overpredict mixture fraction slightly, and those from Zurbach et al. underpredict it. For $r/R > 3$, both give good agreement. The JPDF-ISAT calculations show comparable agreement in the region $r/R < 3$ but not in the outer region. For $r/R > 3$, the experimental data indicate that the mixture fraction approaches zero in the coflow much sooner than the JPDF calculations indicate. This deficiency in the model is discussed in the next subsection.

Fuel, Oxygen, and Temperature

Methane, oxygen, and temperature profiles are plotted in Fig. 2. Consistent with the JPDF–ISAT prediction of mixture fraction, CH_4 is somewhat overpredicted and O_2 underpredicted on the coflow side. Temperature is overpredicted throughout;

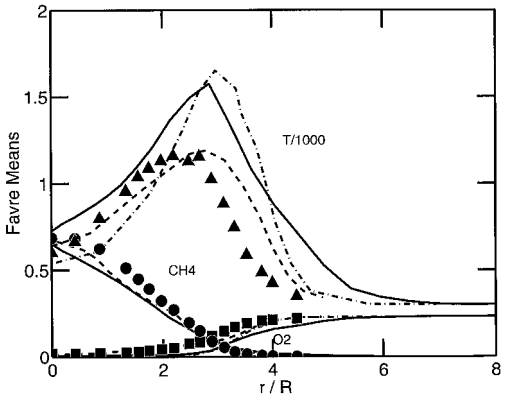

FIG. 2. Radial profiles of Favre-mean concentrations of fuel, oxidizer, and temperature at $x/R = 40$. For legends, see Fig. 1.

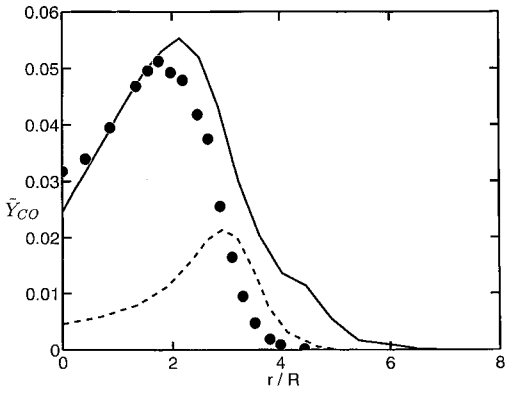

FIG. 4. Minor species: Radial profile of Favre-mean CO concentration at $x/R = 40$. For legends, see Fig. 1.

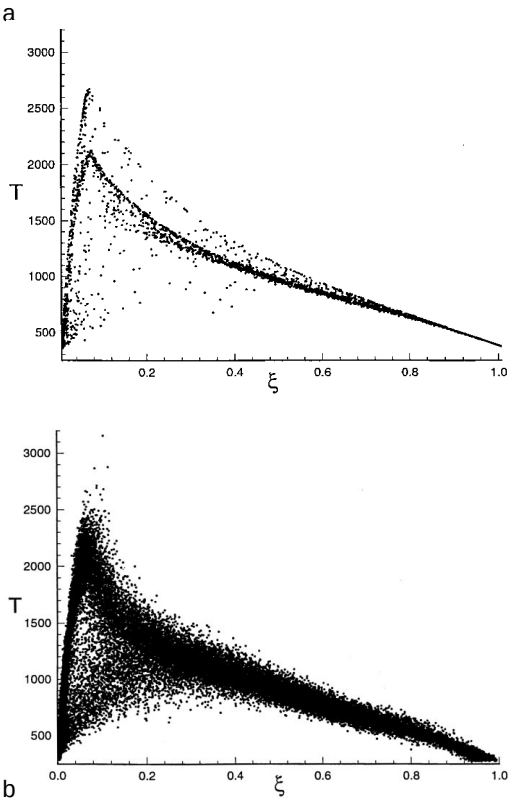

FIG. 3. (a) Scatter plot from JPDF calculations. (b) Scatter plot from experimental data.

more so on the outside. Owing to the better agreement between the calculated mixture fractions by the other two studies with the data in the outer region, their predictions for fuel, oxygen, and temperature in this region are better.

Based on the small value of the stoichiometric mixture fraction ($\xi_s \approx 0.055 = 1/18$), it can be estimated that a 1% overprediction in mixture fraction for lean mixture leads to an 18% underprediction in O_2. This extreme sensitivity to the errors in the calculated mixture fraction profile are evident in the JPDF calculations of O_2 shown in Fig. 2.

The inaccuracy of the mixture fraction profile at the edge of the flame is clearly a deficiency of the model that needs to be rectified. Earlier PDF calculations [5], which used a different algorithm and a different turbulent frequency model, did not exhibit this deficiency. In recent calculations of the turbulent temporal mixing layer, Van Slooten et al. [22] observed a similar problem and ascribed it to the neglect of the pressure transport. A model for the pressure transport was proposed and shown to lead to an improved representation of the edge of the flow. An investigation of this pressure transport model for this flame is an obvious topic for future investigation.

Scatter Plot of T vs. ξ

From the JPDF simulation, about 10,000 particles from the entire domain are uniformly sampled and plotted in $\xi - T$ space in Fig. 3a. Raw data from the experiment, involving 40,000 samples, are plotted in Fig. 3b. As expected, the scatter plot from our calculations clearly shows two distinct curves. The points on the curve corresponding to higher temperature are from the pilot (which is fully burned C_2H_2, H_2, and air). The two scatter plots exhibit similar variations in this space, but there is substantially larger scatter in the data. It is not clear whether

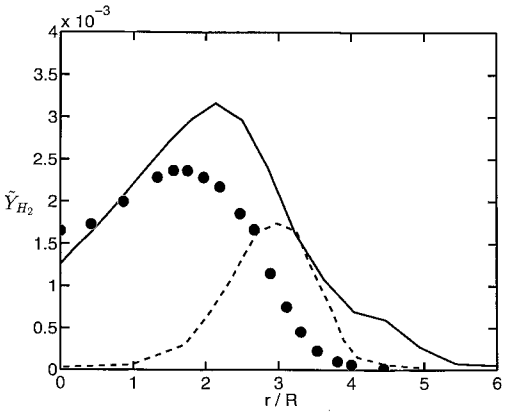

FIG. 5. Intermediate minor species: Radial profile of Favre-mean H_2 concentration at $x/R = 40$. For legends, see Fig. 1.

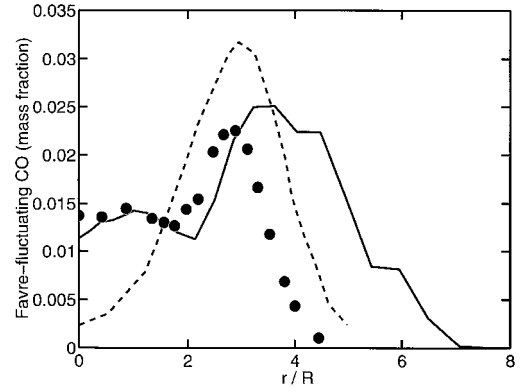

FIG. 6. Radial distribution of $\widetilde{Y''^2_{CO}}^{1/2}$ at $x/R = 40$. For legends, see Fig. 1.

this scatter in the data is due to turbulence (e.g., conditions close to local extinction) or due to experimental errors.

CO

Profiles of CO are shown in Fig. 4. Chen et al. [4] presented scatter plots for CO and concluded that they underpredicted it considerably. Neither they nor Masri and Pope presented radial profiles for CO. The results of Zurbach et al. [6] underpredict CO by a factor of 3 to 5 on the rich side in Fig. 4. The remarkable agreement of the JPDF–ISAT predictions for CO mass fraction with data in this figure, keeping in mind the remarks about the inadequacy of the turbulence model in the outer region, clearly shows the superiority of the detailed chemistry mechanism in modeling this important species.

H_2

Results are also presented in Fig. 5 for an intermediate minor species H_2 whose mean value can be an order of magnitude or more smaller than that of CO. The JPDF–ISAT results are within 25% of the data on the rich side, where the results of Zurbach et al. [6] underpredict H_2 quite significantly. It appears that a three-dimensional ILDM table does not adequately represent the minor species.

Fluctuating CO

The Favre-fluctuating component of CO mass fraction is presented in Fig. 6. Once again, our results from the JPDF–ISAT simulation are substantially better in the central region. The almost flat distribution up to $r/R = 2$ and then the rapid increase in CO fluctuations is captured by the JPDF calculations, in contrast to those of Zurbach et al. [6]. It is also interesting to note that the fluctuating concentrations of CO are comparable in magnitude to the mean CO concentrations.

Conclusions

- A deficiency of the turbulence model used in the JPDF calculations results in excessive spreading in the outer region of the flame. A pressure transport term will be investigated as a possible remedy to this problem.
- The results presented here demonstrate that JPDF–ISAT calculations incorporating detailed chemistry show some significant improvements compared to previous model calculations, particularly for minor species.
- The ISAT algorithm allows us to accomplish efficiently the formidable task of including detailed chemistry in JPDF simulations, while keeping the errors in chemistry calculations within acceptable specified tolerance. The results presented here are from a simulation where chemistry calculations only took 60% of the total CPU time.

Acknowledgments

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COMMENTS

C. H. Priddin, Rolls Royce plc, UK. You mentioned you start your computation with an empty table. Comment on what situations you might want to reuse an existing table.

Author's Reply. The Isat Table is output so that it can be reused in subsequent computations. This saves computational expense if the subsequent computations access the same region of the composition space. If a different region of the composition space is accessed, it is preferable to generate a table from scratch.

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George Kosàly, University of Washington, USA. The deviation between the measured and predicted scalar rms you were showing could well be a major cause of difficulty with NO. Comment on the validity of the turbulence model at 10–20 diameters away from the nozzle.

Author's Reply. Several previous studies with k-ε and Reynolds' stress models have shown that these turbulence models are adequate for piloted jet flames. Apart from the neglect of pressure transport (which is discussed in the paper), the validity of the turbulence modeling aspect of the PDF method is not a concern.

The present work does not consider NO. This will be addressed in future computations of the CH_4/O_2 flames studied recently at Sandia.