NUMERICAL SIMULATION OF TURBULENT NON-EQUILIBRIUM METHANE-AIR JET FLAMES USING MONTE CARLO PDF METHOD

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ABSTRACT

This work presents numerical simulations of a turbulent piloted jet diffusion flame at two Reynolds numbers. Methane partially premixed with air (25% vol. CH_4, 75% vol. air) serves as fuel. The focus is on the investigation of different chemical reaction mechanisms using the Monte Carlo PDF method. This approach has proven to be suitable for the representation of finite-rate chemistry. Hence, a conventionally reduced four step mechanism and a description via two-dimensional manifolds are used as chemical mechanisms. Turbulence is modeled with an eddy-viscosity model. Results shown as first and second moments of velocity are compared with LDV measurements. Species concentrations are compared to data obtained from Raman/Rayleigh/LIF spectroscopy. Even for the flame being close to extinction at high Reynolds number a stable solution can be obtained and the essential features of the flame are well reproduced. The major species and temperature are well predicted whereas the minor species like CO require a more complex chemistry as given by the four step mechanism.

INTRODUCTION

The numerical description of turbulent reactive flows including minor species concentrations has been subject of intensive research during the past decade (Peter, 1984, Pope, 1985, Bilger, 1993, Chen and Chang, 1996, Pitsch *et al.*, 1998). In particular, the modeling of non-equilibrium chemistry gains more and more interest because related effects such as local extinction cannot be captured by fast chemistry approximations (Norris and Pope, 1995, Pope, 1997). Efforts have been put forward to apply more sophisticated chemical mechanisms (Pope, 1997, Sung *et al.*, 1998, Chen, 1997). For technical applications, however, the emphasis is on a reasonable accuracy at moderate costs. Hence, a compromise has to be found where a sufficiently complex mechanism yields reasonable results in particular for minor species. This paper focuses on an elaborated description of non-premixed combustion while the aspects of turbulence modeling are treated with an eddy-viscosity model (Jones and Launder, 1972, Launder *et al.*, 1972).

Two different chemical mechanisms are applied in this work, namely an Intrinsic Low-Dimensional Manifolds (ILDM) mechanism (Maas and Pope, 1992a, 1992b) and a reduced four step mechanism (Yuasa and Chen, 1999). The complete model consists of a combined Monte Carlo - finite-volume method (Correa *et al.*, 1994) where chemical reactions are treated by the subcode solving for the joint composition probability density function (PDF) and the flow field by the computational fluid dynamics (CFD) code.

At the *Turbulent Non-Premixed Flames* Workshop (TNF, 1998) a set of task flames has been established. A series of partially-premixed methane-air flames has been investigated experimentally and serves as reference case (Barlow and Frank, 1998). Two flames out of this series at Reynolds numbers of $22,400$ and $44,800$, respectively, are subject of the current investigation.

APPLIED MODELS

In order to describe the chemical reactions in closed form, the transport equation of the joint composition PDF is solved. Since finite-difference methods for the discretization are not feasible for more than two scalars, a Monte Carlo method is used to solve for the joint PDF where ensembles of notional particles with a set of specified scalars represent the PDF in discrete form. Given the scalars of a single particle, the exact position in

composition space is determined and hence the chemical source term for each property can be evaluated. The transport equation for the mass weighted joint PDF $\tilde{P}_\phi$ is given by

$$\overline{\rho}\frac{\partial \tilde{P}_\phi(\boldsymbol{\psi};\mathbf{x},t)}{\partial t} + \overline{\rho}\tilde{u}_j\frac{\partial \tilde{P}_\phi(\boldsymbol{\psi};\mathbf{x},t)}{\partial x_j} = -\frac{\partial}{\partial \psi_\alpha}\left[\overline{\rho}S_\alpha \tilde{P}_\phi(\boldsymbol{\psi};\mathbf{x},t)\right] - \frac{\partial}{\partial x_j}\left[\overline{\rho}\langle u_j''|\boldsymbol{\phi}=\boldsymbol{\psi}\rangle \tilde{P}_\phi(\boldsymbol{\psi};\mathbf{x},t)\right] - \frac{\partial}{\partial \psi_\alpha}\left[\langle -\frac{\partial J_k^\alpha}{\partial x_k}|\boldsymbol{\phi}=\boldsymbol{\psi}\rangle \tilde{P}_\phi(\boldsymbol{\psi};\mathbf{x},t)\right] \quad (1)$$

where the expression $\langle a|b\rangle$ denotes the mean of a conditioned on the event b. In this equation $\overline{\rho}$, $\tilde{u}_j$, and u_j'' are the mean density, the Favre-averaged velocity and its fluctuation. The scalar ϕ_α denotes a random variable that can assume certain values ψ_α in sample space. All N_α variables together represent the current state $\boldsymbol{\phi}$ in composition space. The terms on the left hand side need no modeling and likewise the first term on the right hand side emphasizing the closed description of the chemical source term S_α. The remaining terms represent turbulent diffusion in physical space and diffusion due to molecular mixing in composition space. They both require modeling.

Employing an Eulerian description for the solution procedure in the Monte Carlo subcode, the convective and turbulent diffusive transport across cell surfaces are carried out through a finite-volume discretization of the PDF transport equation in physical space: The transport process is simulated by randomly selecting particles of the respective neighbor cells and copying their scalars onto the particles of the cell of consideration. Hence, the number of particles remains constant in each cell throughout the simulation. The mixing process and chemical reactions in scalar space are simulated for each single cell without interaction with the neighbors. By undergoing theses processes, every particle representing a state in composition space moves to some new position.

Since the velocity is not solved within the PDF approach, a coupling between the code solving for the joint composition PDF and a finite-volume code is established (Correa *et al.*, 1994).

Within the frame of Favre-averaging, the conservation equation of mass yields

$$\frac{\partial \overline{\rho}}{\partial t} + \frac{\partial(\overline{\rho}\tilde{u}_i)}{\partial x_i} = 0 \quad (2)$$

and the transport equation for momentum reads

$$\overline{\rho}\frac{\partial \tilde{u}_i}{\partial t} + \overline{\rho}\tilde{u}_j\frac{\partial \tilde{u}_i}{\partial x_j} = -\frac{\partial \overline{p}}{\partial x_i} + \overline{\rho}g_i + \frac{\partial}{\partial x_j}\left[\nu\overline{\rho}\left(\frac{\partial \tilde{u}_i}{\partial x_j}+\frac{\partial \tilde{u}_j}{\partial x_i}\right) - \overline{\rho}\widetilde{u_i''u_j''}\right] \quad (3)$$

The solution procedure bases on an iterative process of exchanging information: The CFD code passes the velocity field and turbulent quantities to the Monte Carlo subcode. There the solution of the PDF transport equation proceeds in time. Then the mean density is evaluated through an ensemble averaging process and fed back to the CFD code because chemical reactions mainly influence the flow field through the mean density. The impact of the laminar diffusion coefficient becomes negligible for sufficiently high Reynolds numbers.

Turbulence Model

The Reynolds stresses are closed with an eddy-viscosity approximation

$$\widetilde{u_i''u_j''} = -\nu_t\left(\frac{\partial \tilde{u}_i}{\partial x_j}+\frac{\partial \tilde{u}_j}{\partial x_i}-\frac{2}{3}\delta_{ij}\frac{\partial \tilde{u}_k}{\partial x_k}\right)+\frac{2}{3}\delta_{ij}\tilde{k} \quad (4)$$

with $\tilde{k}$ being the turbulent kinetic energy $\frac{1}{2}\widetilde{u_k''u_k''}$. The eddy viscosity ν_t is defined as $\nu_t = C_\mu\tilde{k}^2/\tilde{\varepsilon}$ where $\tilde{\varepsilon}$ denotes the dissipation rate. In contrast to the standard $k-\varepsilon$ model (Jones and Launder, 1972), a modified version suggested by Launder *et al.* (1972) is used. The constants σ_k and σ_ε take standard values of 1.0 and 1.3, respectively, and $C_{\varepsilon 1} = 1.44$. The coefficients C_μ and $C_{\varepsilon 2}$ are now functions of the centerline decay of the mean axial velocity according to

$$C_\mu = 0.09 - 0.04F_{jet}\,,\; C_{\varepsilon 2} = 1.92 - 0.0667F_{jet} \quad (5)$$

where F_{jet} is a function of the decay of the centerline velocity.

Chemistry Model

The chemical source S_α is given by the first term on the right hand side of equation (1) and is determined through a reaction mechanism. Two different chemical models are applied and compared. First, the ILDM method (Maas and Pope, 1992a, 1992b) is used. The low-dimensional manifolds are parameterized by the mixture fraction f, assuming equal diffusivity, and the mass fractions Y_{CO_2} and Y_{H_2O} as reaction progress variables. These three scalars define the current state on a low-dimensional manifold in composition space.

The question arising in terms of technical applications is whether this ILDM description is comparable to an advanced higher-order reduced mechanism. Therefore, a four step mechanism is applied as alternative chemistry model. This mechanism has been derived by Chen and the parameterization has been modified and improved by Yuasa (Yuasa and Chen, 1999). The mechanism reads

$$\frac{1}{4}O_2 + H = \frac{1}{2}H_2O \quad (6)$$

$$H_2 + \frac{1}{4}O_2 = H + \frac{1}{2}H_2O \quad (7)$$

$$\frac{1}{4}O_2 + CO + \frac{1}{2}H_2O = H + CO_2 \quad (8)$$

$$CH_4 + \frac{3}{4}O_2 + H = 2H_2 + CO + \frac{1}{2}H_2O \quad (9)$$

and its evaluation is also based on the assumption of equal molecular diffusivity. Hence, the current state in composition space is determined by the mixture fraction and four reaction progress variables, namely the mass fractions Y_{CH_4}, Y_{CO}, Y_{O_2}, and Y_H.

Since chemical reaction mechanisms obey a stiff system of differential equations, an online integration is not feasible for most applications. Therefore, for both, the ILDM method and the reduced mechanism, look-up tables consisting of pre-integrated source terms as a function of the current state in composition space are generated. Given a set of scalars and a time increment, the new state can be evaluated via a multi-dimensional interpolation of the pre-integrated source terms. Similarly, other properties such as temperature or density can be determined.

Turbulent Diffusion Model

The conditional expectation of the velocity fluctuations $\langle u_j'' | \boldsymbol{\phi} = \boldsymbol{\psi} \rangle$ given in the second term on the right hand side of equation (1) needs to be modeled. It represents the diffusive transport process in physical space due to turbulent fluctuations. It is modeled using a gradient diffusion approximation according to

$$\langle u_j'' | \boldsymbol{\phi} = \boldsymbol{\psi} \rangle \tilde{P}_\phi = -\frac{\nu_t}{\sigma_P} \frac{\partial \tilde{P}_\phi}{\partial x_j} \tag{10}$$

with a turbulent Prandtl number $\sigma_P = 0.85$.

Mixing Model

In this work, the *Modified Curl's* model (Janicka *et al.*, 1979) is applied for the molecular mixing in composition space $\boldsymbol{\psi}$ represented by the third term on the right hand side of equation (1). It reads

$$-\frac{\partial}{\partial \psi_\alpha}\left[\langle -\frac{\partial J_k^\alpha}{\partial x_k} | \boldsymbol{\phi} = \boldsymbol{\psi} \rangle \tilde{P}_\phi(\boldsymbol{\psi}; \mathbf{x}, t)\right] = -\frac{\partial}{\partial \psi_\alpha}\left[\beta M(\psi_\alpha) \tilde{P}_\phi(\boldsymbol{\psi}; \mathbf{x}, t)\right] \tag{11}$$

where $\beta = 3$ and

$$M(\psi_\alpha) = \int \tilde{P}_\phi(\hat{\phi}_\alpha)\, \tilde{P}_\phi(\check{\phi}_\alpha) H(\hat{\phi}_\alpha, \check{\phi}_\alpha, \psi_\alpha)\, d\hat{\phi}_\alpha\, d\check{\phi}_\alpha \tag{12}$$

with

$$H(\hat{\phi}_\alpha, \check{\phi}_\alpha, \psi_\alpha) = \begin{cases} \frac{1}{|\hat{\phi}_\alpha - \check{\phi}_\alpha|} & \text{for } \psi_\alpha \in [\hat{\phi}_\alpha, \check{\phi}_\alpha] \\ 0 & \text{otherwise} \end{cases} \tag{13}$$

EXPERIMENTAL SYSTEM

The flow configuration consists of a turbulent, piloted jet flame with a nozzle diameter $d = 7.2$ mm (TNF, 1998). Methane partially premixed with air (25 % vol. CH_4, 75 % vol. air) serves as fuel. The measurements show that the strain rates are sufficiently high that the flame can be treated as a diffusion flame. The flame stabilization is guaranteed by an ensemble of tiny flames located around the burner being approximated as a ring-shaped burner. This system is embedded in co-flowing air. The composition of the pilot stream is such that the mixing processes of the system can be treated with only one conserved scalar for the elements and also for the enthalpy with an error less than 5 %. A series of six flames at increasing Reynolds numbers have been extensively investigated using Raman / Rayleigh / Laser Induced Fluorescence (LIF) spectroscopy (Barlow and Frank, 1998). Two configurations at a Reynolds number of 22,400 (Flame D) and 44,800 (Flame F) are subject of the current work. In particular Flame F shows a significant amount of local extinction. The velocity fields of both flames have been measured using Laser Doppler Velocimetry (LDV).

NUMERICAL SETUP

Discretization and Solution Method

The computations are carried out on a rectangular staggered grid condensed near the burner. Assuming axisymmetry, the equations are discretized in physical space using the finite-volume method at a resolution of 80×70 grid points in axial and radial direction, respectively. Computations on a 120×80 grid showed similar results within the statistical fluctuations being inherent for this stochastic solution method.

The flow field is solved using a two-dimensional elliptic finite-volume CFD code employing the SIMPLE algorithm (Patankar and Spalding, 1972) to ensure the conservation of mass and momentum.

For the Monte Carlo PDF calculation 100 particles are used in each cell for the simulations with ILDM chemistry and 50 particles for reduced four step mechanism. Whereas the remaining variables throughout the simulation require about 5 MB RAM the size of the look-up table for the ILDM method is about 25 MB and for the four step mechanism about 160 MB. The CPU time required for one run using ILDM chemistry is in the range of 60 hours on an ALPHA LX533 Linux-workstation.

Initial and Boundary Conditions

At the inlet boundary, the velocity and turbulent quantities are prescribed according to LDV measurements and the dissipation rate is prescribed according to a proposed formula by Masri and Pope (1990). The boundary at the outer radius represents the quiescent surrounding providing fluid for the entrainment. The initial conditions are block profiles through the entire computational domain according to the inlet conditions.

RESULTS AND DISCUSSION

In the first part of the discussion, the focus is on results of the simulation using ILDM chemistry. Computations with the reduced four step mechanism give comparable results for velocity and major species and,

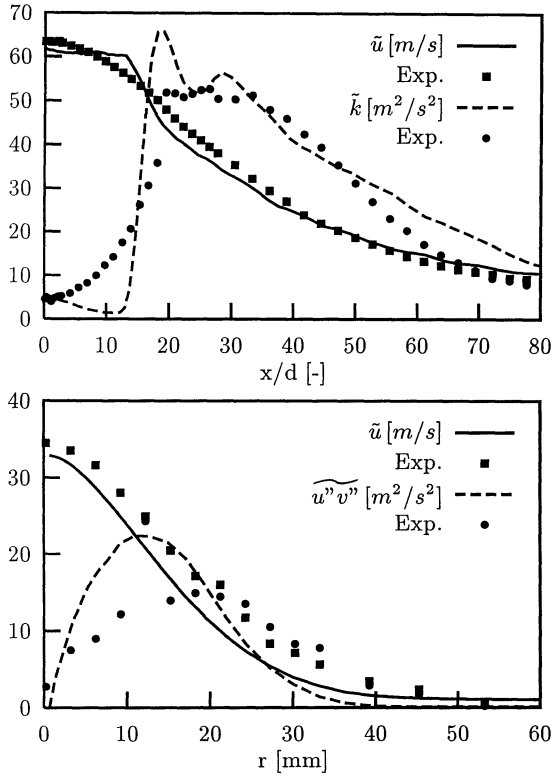

Figure 1: Flame D/ILDM: Axial distribution of mean velocity and mean turbulent kinetic energy (top). Radial distribution of mean axial velocity and shear stress at $x/d = 30$ (bottom).

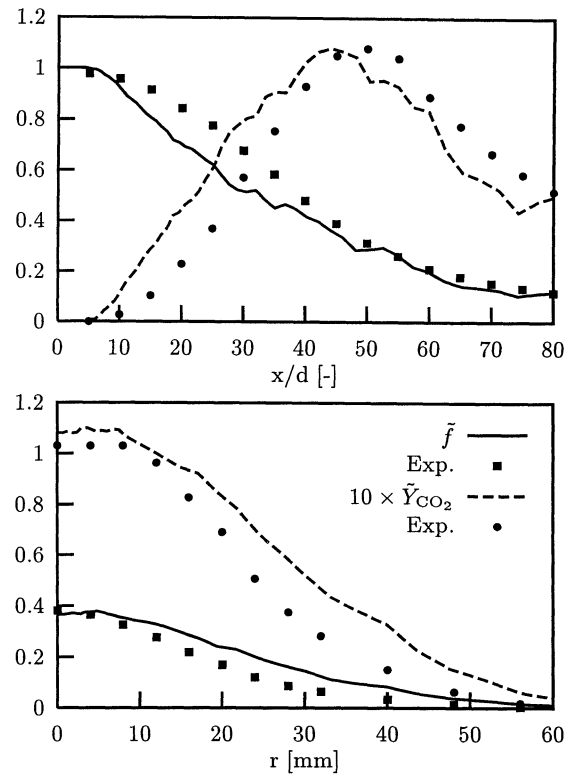

Figure 2: Flame D/ILDM: Axial and radial ($x/d = 45$) distribution of mixture fraction and mass fraction of CO_2.

hence, are not shown here.

The flow field of Flame D can be captured with good accuracy using the modified $k-\varepsilon$ model. Although the pilot might have an impact on the near field, the shear flow character of the flow remains. Profiles of mean axial velocity are shown along the axis at $x/d = 30$ together with the turbulent kinetic energy and the shear stress component $\widetilde{u''v''}$ (Fig. 1). Besides the deviations, it can be concluded that the prediction of flow field is sufficient for further investigations on the scalar field, especially with respect to chemical reactions. For more accurate simulations, a Reynolds stress closure could be considered as shown by Jones and Kakhi (1998).

The mean values of mixture fraction f and mass fraction of CO_2 as representative of the major species are shown along the axis and at the downstream locations $x/d = 45$ (Fig. 2). The axial profiles show good agreement and, hence, Y_{CO_2} matches the peak values at $f_{st} = 0.351$ at a downstream location of $x/d \approx 45$. The radial profiles reveal an over-prediction of the spreading rate of the scalars. In order to circumvent this deficiency, the species concentrations are better examined in terms of their means conditioned on the mixture fraction. The conditional means of CO_2 and the temperature given in this representation in figure 3 reveal that the ILDM mechanism gives a good description of the major species.

Another interesting effect is local extinction in turbulent flames at high Reynolds numbers. Experimental data given in scatter plots show extinguished samples. The Monte Carlo PDF method also provides the possibility to generate scatter plots and especially Flame F exhibits a significant amount of local extinction. In figure 4 showing the conditional temperature at the downstream location $x/D = 30$ it is confirmed that the simulations applying the ILDM mechanism can capture this effect although not to the extent of the experiments.

Predictions of minor species distributions must be attributed to the chemistry model used in the simulation. To further investigate this aspect, a numerical simulation using the reduced four step mechanism is carried out. To discuss the performance of the chemistry models, the conditional means and variances of CO are shown for Flame D and Flame F at downstream location $x/d = 45$ (Fig. 5).

The ILDM chemistry shows a very good performance on the lean side but fails sharply beyond f_{st}. The explanation can be found in the derivation of the ILDM method which de-couples *fast* and *slow* chemical time scales. This approach fails if the number of characteristic time scales of the chemistry being in the same

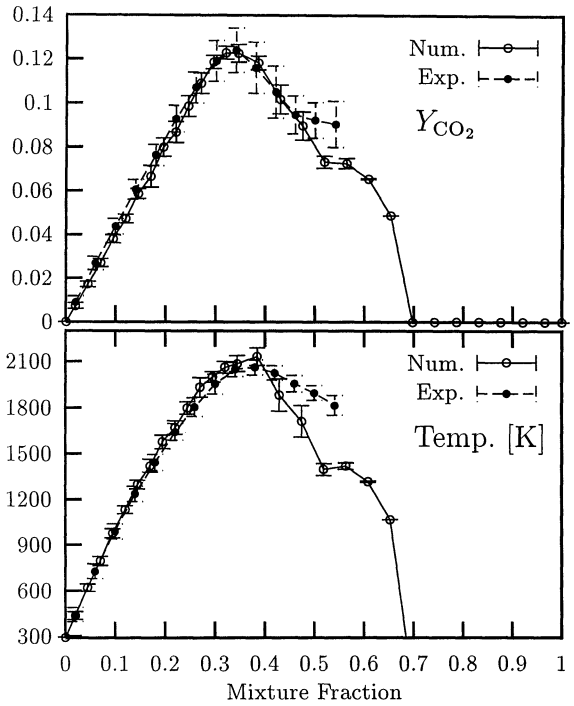

Figure 3: Flame D/ILDM: Conditional mean of mass fraction of CO_2 (top) and temperature (bottom) at $x/D = 45$.

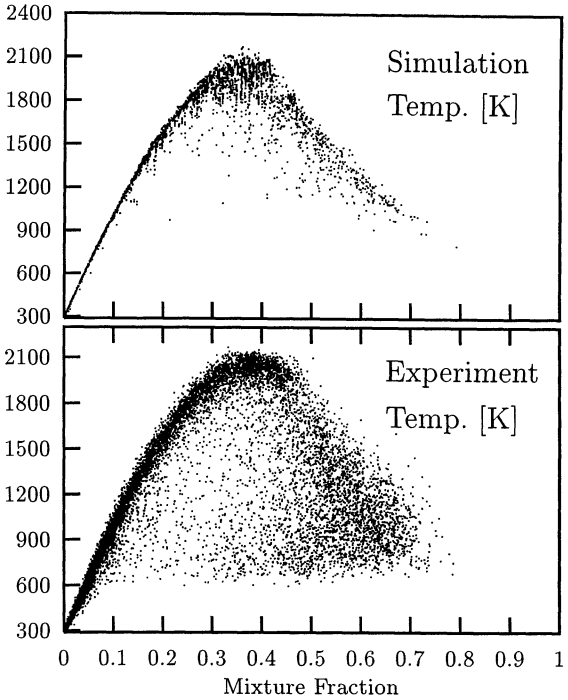

Figure 4: Flame F/ILDM: Scatter plots of temperature at $x/D = 30$

order of magnitude becomes larger than the number of variables chosen to parameterize the low-dimensional manifold. It can be assumed that this is the case on the fuel rich side and here at least a third eigenvalue or characteristic time scale, respectively, needs to be considered.

The reduced four step mechanism on the other hand leads to CO predictions that reach much further into the rich flame zone although the high concentration of 9% in Flame D is not reached.

It should also be noted that the conditional variances of CO are under-predicted. The explanation for this feature is the difficulty to obtain a stably burning flame for the reduced four step mechanism. Both, Flame D and Flame F, need to be stabilized by employing ILDM chemistry and perfect mixing in scalar space in the near field for $x/D < 7.5$. Further downstream where the reduced four step chemistry is applied, enhanced mixing is necessary to keep the flame stably burning. The number of pairs involved in the mixing process is determined by $N_{pairs} = \beta \Delta t N_{total}(\tilde{\varepsilon}/\tilde{k})$ where N_{total} is the number of particle in the cell and $\beta = 3$ (see sect. Mixing Model). For this simulation the number of pairs is determined by $N_{pairs} = \min[10 \times \beta \Delta t N_{total}(\tilde{\varepsilon}/\tilde{k}), N_{total}]$. It is assumed that the results further downstream are characteristic for the reduced four step mechanism and the impact of ILDM chemistry becomes negligible.

It can be concluded that the reduced four step mechanism is indeed sufficient to give a reasonable to good prediction of CO but the advantage of a closed description of the chemical source term in the PDF transport equation cannot be entirely appreciated because the sensitive mixing term needs to be modeled.

SUMMARY AND CONCLUSION

Simulations of a turbulent piloted methane-air jet at Reynolds number of 22,400 and 44,800 using Monte Carlo PDF methods have been carried out. Two different chemistry mechanisms have been used, namely a description based on the ILDM method with two reaction progress variables and reduced four step mechanism. It has been shown on the basis of a reliable set of experimental data and a representation in mixture fraction space that a two dimensional manifold is insufficient to describe the reactions on the rich side of the flame to correctly predict the CO concentration. With the four step mechanism, results could have been improved but the numerical treatment in terms of artificial stabilization and tabulation is more difficult.

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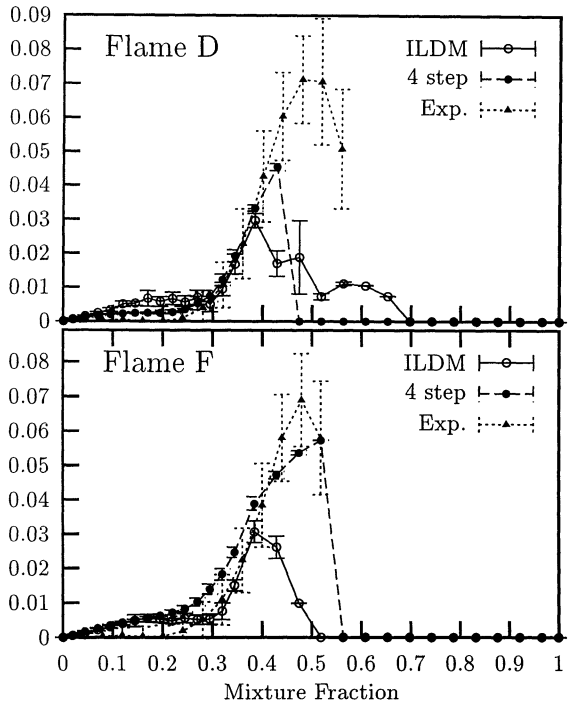

Figure 5: Conditional mean of mass fraction of CO using ILDM chemistry and the reduced four step mechanism for Flame D (top) and Flame F (bottom) at $x/d = 45$.

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