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NO Predictions in Methane-Air Jet Flames Using the Conditional Moment Closure Method

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ABSTRACT

NO predictions for turbulent piloted methane-air (1:3) jet flames with a Reynolds number of ~22400 using Conditional Moment Closure (CMC) method are presented. GRI-Mech 2.11 is employed to represent the chemistry. Predictions are compared with the multiscalar Raman/Rayleigh/LIF measurements of Barlow and Frank (1997) and LDV two-component velocity measurements of Schneider et al. (1998).

The flow and mixing field results are in a fair quantitative agreement with the measurements. The mixing rates near the nozzle are somewhat over-predicted and the predicted flame is slightly shorter than reported in the experiment. A reasonable agreement for predicted NO levels is observed. The NO predictions are over-predicted by ~50% on the fuel-lean side and under-predicted by ~20% on the fuel-rich side. At downstream locations, the overall trends of predictions are similar to the upstream ones and show even better quantitative agreement with the measurement.

INTRODUCTION

The need for reduction of pollutant emissions is forcing changes in combustion technology at an ever increasing rate. Computational fluid dynamics (CFD) codes are widely used in the flow and mixing design of combustors resulting in much reduced test-rig development times. Engineering capability for predicting emissions such as carbon monoxide, CO, nitric oxide, NO, soot and polycyclic aromatic hydrocarbons (PCAH) is quite inadequate, however. This is due in part to the need for the use of complex chemical mechanisms for the prediction of these pollutants, making computation a mammoth task even in two-dimensional laminar flow (Smooke et al., 1996), and in part due to the difficulties associated with handling the interactions between the chemistry and the turbulence.

Conditional Moment Closure (CMC) (Klimenko, 1990, Bilger, 1993) is a method for handling turbulence chemistry interactions which is capable of being used with large chemical mechanisms at modest computational cost. The basis of the method is the idea that most of the fluctuation in temperature and composition can be associated with one variable and conditional averaging with respect to that variable allows closure of the conditional average chemical reaction rate terms. For the nonpremixed combustion systems considered here, the conditioning variable of choice is the mixture fraction. This is defined as the mass fraction of an inert tracer entering with the fuel, normalised to be unity in the unmixed fuel and zero in the unmixed oxidant.

Excellent results have been obtained for NO predictions in turbulent jet diffusion flames of hydrogen (Smith et al., 1992, 1995). Here we apply the method to predictions for a turbulent diffusion flame formed from a partially premixed jet of methane and air. Results are compared with the laser measurements of Barlow and Frank (1997) and Schneider et al. (1998).

GOVERNING EQUATIONS AND METHOD OF SOLUTION

The governing equations describing the problem are two-fold: for the flow and mixing field and for the reactive scalar field. The governing equations for the flow and mixing field for the case under study may be expressed by the usual Favre-averaged equations in axisymmetric boundary-layer form for continuity, momentum, turbulence kinetic energy, turbulent kinetic energy dissipation rate and the mean and variance of the mixture fraction. The closure used here for the turbulence is the k-ε-g model of Launder et al. (1972). The governing equations for the reactive scalars are obtained by implementation of the CMC method to a steady axisymmetric jet flame. Experimental results show that in such flows there is little cross-stream variation of the conditional averages across the flow (Bilger, 1993). This weak cross-stream dependence of the conditional averages is supported by asymptotic analysis of Klimenko (1995). Using this result the CMC equation can be brought into cross-stream averaged form by integrating across the flow (Klimenko, 1990), giving

$$U^* \frac{\partial Q_i}{\partial x} = \langle W_i | \eta \rangle + N^* \frac{\partial^2 Q_i}{\partial \eta^2}, \qquad (1)$$

where

$$\begin{aligned} U^* &\equiv \frac{1}{\langle \rho | \eta \rangle P^*} \int_0^R \bar{\rho} \langle u | \eta \rangle \tilde{P}_\eta \, r dr, \\ N^* &\equiv \frac{1}{\langle \rho | \eta \rangle P^*} \int_0^R \bar{\rho} \langle N | \eta \rangle \tilde{P}_\eta r \, dr, \\ P^* &\equiv \frac{1}{\langle \rho | \eta \rangle} \int_0^R \bar{\rho} \tilde{P}_\eta r \, dr. \end{aligned} \qquad (2)$$

Q_i is the conditional average of reactive scalar i, defined as $Q_i(\eta; x) \equiv \langle Y_i(x,t) | \xi(x,t) = \eta \rangle$, W_i is the rate of chemical production of species i per unit mass of mixture,

$N \equiv D\nabla\xi \cdot \nabla\xi$ is the scalar dissipation rate (D represents molecular diffusivity) and η is the nominated value of mixture fraction, ξ. The angle brackets denote ensemble averaging subject to the condition to the right of the vertical bar. The asterisk indicates that the quantity is weighted with the local pdf of the mixture fraction, P, and integrated across the flow. The straight overbar denotes a conventional average and the tilde represents a Favre average. x and r are the axial and radial coordinates respectively. u is the axial component of the velocity, ρ is density and R is the bounding radius, assumed sufficiently large so that the integrals converge. The enthalpy equation has the same form as equation (1), with the chemical production term replaced with the radiation sink term.

The local pdf of mixture fraction and as a result P^* are obtained from the flow and mixing field results using a presumed form for the pdf. The cross-stream averaged conditional scalar dissipation, N^*, can be evaluated from the cross-stream integral of the pdf transport equation. The pdf transport equation is an adjoint equation to the CMC equation and its use ensures conservation of species. The cross-stream averaging of the pdf transport equation relates the second derivative of conditional mean scalar dissipation to the conditional mean mass flux in form of

$$N^* P^* = -\int_0^{\eta} \int_0^{\eta'} \frac{\partial}{\partial x}\left(U^* P^*\right) d\eta'' \, d\eta' \cdot \qquad (3)$$

The flow and mixing field and reactive scalar field are related through the mean density field. At each axial step, the flow and mixing field information obtained from the CFD code are passed to the CMC code. In turn, the CMC code solves for the species mass fractions and enthalpy and passes information on the density to the CFD code. This routine is repeated as many times as necessary until the whole computational domain is computed. In practice, many steps are taken in the CMC code for each step taken for the CFD code because a relatively stiff system of equations is involved. Mean values are obtained by weighting with the local pdf for the mixture fraction. This is assumed to have a clipped Gaussian form.

EXPERIMENTAL DATA

Raman/Rayleigh/LIF measurements of temperature, mixture fraction, N_2, O_2, H_2O, H_2, CH_4, CO, CO_2, OH and NO, made by Barlow and Frank (1997) and velocity measurements made by Schneider et al. (1998) are used for comparison with the predictions. The axial and radial profiles of Favre averaged values and root mean square (rms) of fluctuations along with the conditional averages and rms of conditional fluctuations are reported at various axial locations.

The burner is an axisymmetric jet with a jet nozzle diameter of 7.2 mm and an outer annulus diameter of the pilot of 18.2 mm, centred in a stream of co-flowing air. The jet velocity is 49.6 m/s (±2 m/s) which has a jet Reynolds number of ~22400. The co-flow velocity is 0.9 m/s. The main fuel is a mixture of one part methane and three parts air by volume at temperature of 294 K. The fuel has a stoichiometric mixture fraction, ξ_{st}=0.351.

The pilot burns a lean pre-mixture of C_2H_2, H_2, air, CO_2 and N_2 with equivalence ratio of 0.77. This mixture has the same nominal enthalpy and equilibrium composition as methane-air (1:3). The pilot burnt gas velocity is reported as 11.4 m/s (±0.5 m/s) based on the cold mass flow rate and density at the estimated exit condition (Barlow and Frank, 1997).

RESULTS AND DISCUSSION

The calculations are carried out down to x/D=100. The chemistry is represented by the GRI-Mech 2.11 mechanism. GRI-Mech consists of 279 reactions and involves 49 species and contains C_1 and C_2 reactions along with thermal, prompt and N_2O mechanisms for NO formation. Radiative heat loss is modelled by RADCAL (Sivathanu and Gore, 1993) radiation sub-model. Adiabatic equilibrium compositions are employed for the reactive scalars down to five jet diameters, in order to assure the ignition of the flame in the near-field region due to high mixing rates.

Flow and Mixing Field Results

The quality of flow and mixing field is discussed first to examine how well the flow and mixing field is modelled. Figure 1 shows the axial profiles of Favre averaged velocity and turbulence predictions. The Favre averaged velocity is normalised by centreline mean velocity at the jet exit plane and the root mean square of axial velocity fluctuations, u', is normalised by mean velocity on the centreline.

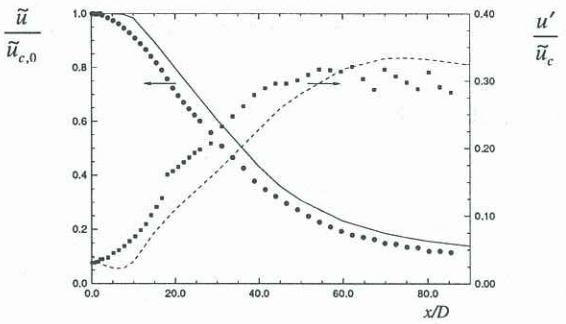

Figure 1: Axial profiles of mean velocity and turbulence where $\tilde{u}$ is the mean axial velocity, u' is the rms of fluctuation of axial velocity, $\tilde{u}_c$ is the mean centerline velocity and $\tilde{u}_{c,0}$ is the centerline velocity at the jet exit. CMC (solid and dashed lines); measurements (symbols).

The LDV measurements of velocity and turbulence made by Schneider et al. (1998) are also plotted in Figure 1. It can be seen that the predictions are in reasonably good agreement with the measurements. The axial profile of Favre averaged mixture fraction and its fluctuations on the centreline are plotted in Figure 2. The mean mixture fraction on the centreline is in good agreement with the measurements of Barlow and Frank (1997). Fluctuations in mixture fraction are somewhat over-predicted particularly for downstream.

The stoichiometric flame length is predicted to be 44 jet diameters and the visible flame length is estimated from the predictions ~64 jet diameters. These are in very good agreement with the measured stoichiometric and visible flame lengths which are reported as 47 and 67 jet diameters, respectively.

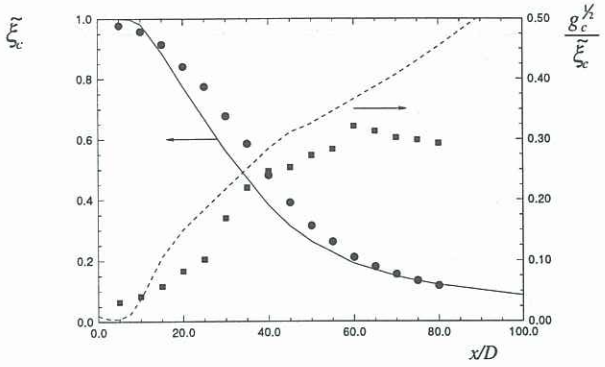

Figure 2: Axial profiles of Favre averaged mixture and its fluctuations on the centreline. $\tilde{\xi}_c$: CMC predictions (line); measurements (circles) ; $g_c^{1/2}/\tilde{\xi}_c$: CMC predictions (dashed line), measurements (squares).

The total enthalpy lost to the surroundings in the computational domain is calculated by subtracting the radiative standardised enthalpy from the adiabatic standardised enthalpy and found to be 2.16 kW. This quantity when normalised by the total power of the flame (17.34 kW) represents the radiant fraction. The radiant fraction is predicted to be ~12%, which is 2.4 times greater than that measured (~5.1%) by radiometers by Barlow and Frank (1997). This is a large discrepancy and there should be a corresponding discrepancy in predicted and measured temperatures.

Reactive Scalar Field Results

Figure 3 shows measured and predicted conditional mean temperature at x/D=30. Predicted temperature levels are in very good agreement with measurements on the fuel-lean side. The predicted and measured peak conditional mean temperatures occur at η=0.38 with values of 2022K and 2058K respectively which shows very good agreement as well. On the fuel-rich side, however, the temperature levels tend to deviate from the measurements and higher levels of temperature (by 10%) are predicted. At x/D=60 the mixture is always fuel lean and the predictions for conditional averaged temperature are about 50K below the measurements. This is only about half of the discrepancy that can be inferred from the discrepancy in the radiation loss reported above.

CMC predictions for reactive species are excellent on the fuel-lean side. Significant deviations from measurements are observed for CH_4, O_2, CO and H_2 on the fuel-rich side (not shown) and these are consistent with those found for temperature. Predicted CO levels on the fuel-lean side show fair agreement with measurement while the predicted peak carbon monoxide value shifts to the rich side of stoichiometric. Levels of H_2 are well predicted on the fuel-lean side but they are over-predicted on the fuel-rich side. The hydroxyl mass fractions are in reasonable agreement with measurements with the peak value, which occurs at stoichiometric, being over-predicted by 20%. It seems that the chemistry is poorly modelled on the fuel-rich side.

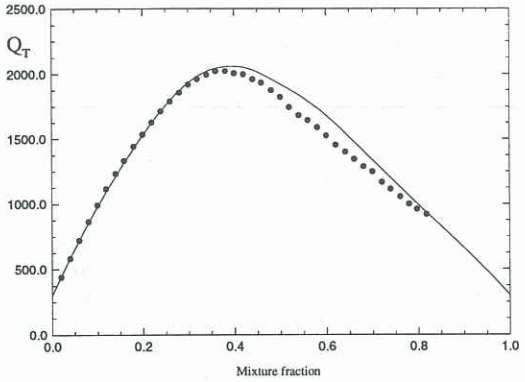

Figure 3: Conditional mean temperature at x/D=30. CMC (line), measurements (circles).

Predicted NO levels are in fair agreement with the measured levels (Figure 4). The predicted NO level peaks at stoichiometric as in the measurements. The predictions are high by ~50% on the fuel-lean side. Predictions show faster consumption of NO on the fuel-rich side and it is under-predicted there by ~20%.

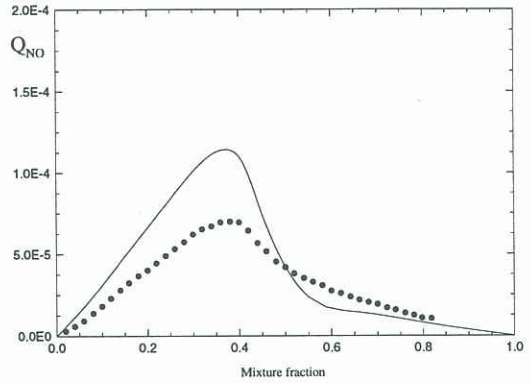

Figure 4: Conditional mean NO mass fractions at x/D=30. CMC predictions (line), measurements (circles).

Smooke et al. (1996) investigate issues related to the computation and measurement of NO in laminar methane-air diffusion flames. They employ both GRI-Mech and a combined mechanism using the C_2 reaction set of Smooke et al. (1992) with the NO_x submechanism of Drake and Blint (1991). The results of their study indicate that significant variations in NO levels are obtained by using different mechanisms. They found that the combined mechanism significantly over-predicts NO levels but that GRI-Mech produces excellent agreement for most of the features of the measured NO with the peak NO mole fractions underpredicted by less than 30% of the experimental value in the opposite direction to our finding. Their temperatures were quite a lot lower than in our flame It is believed that the inadequacy of current chemical kinetics for reaction in nonpremixed flames could be a main cause for the discrepancies found. Other causes could be the need for a higher order closure for

the conditional reaction rate.(Kronenburg, et al., 1998) or numerical errors.

Favre averaged statistics are obtained by the weighting of the conditional mean statistics with the local pdf over the entire mixture fraction space. Therefore, the quality of predictions for Favre averaged statistics are affected by the quality of both the conditional mean statistics and the pdf of mixture fraction, and hence should not exceed the quality of the conditional averaged predictions. Radial profiles of Favre averaged temperature and NO mass fractions at the axial location of 60 jet diameters are plotted in Figure 5. The agreement for temperature is very good and not significant lower than the measurements as the discrepancy in the radiation loss referred to above would imply. In the measurements there must be a small but significant radial variation of conditional average temperatures to account for the lack of discrepancy here that was apparent in the conditional values. The NO predictions are some 50% high.

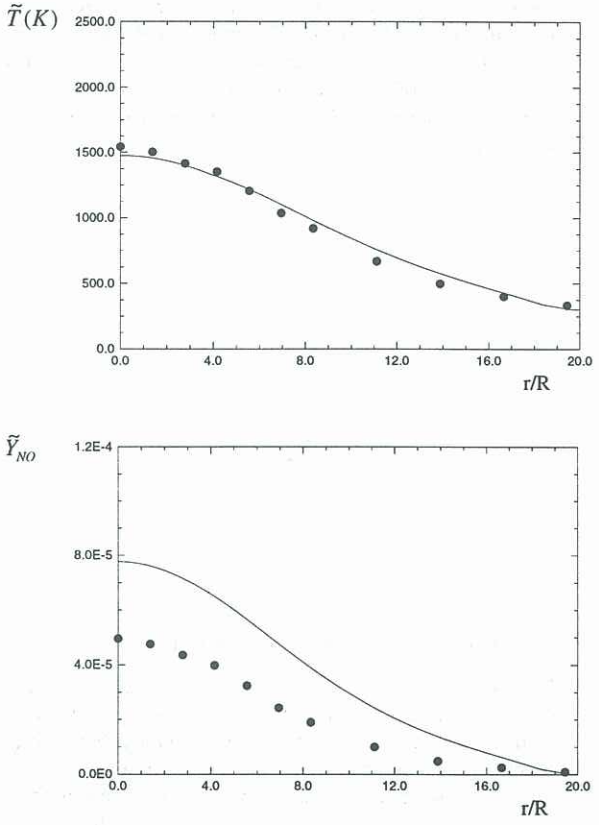

Figure 5: Radial profile of Favre averaged temperature and NO at x/D=60. CMC predictions (line); measurements (circles).

CONCLUSION

The flow and mixing field results are in quite good quantitative agreement with the measurements. The k-ε-g turbulence model somewhat over-predicts the mixing rates near the nozzle and the predicted flame is slightly shorter than that reported in the experiment. Predicted temperature levels are in much better agreement with the measurements than is consistent with the discrepancy in the radiant loss. The radiation measurements need to be checked. Conversion of fuel to intermediates is over-predicted on the rich side of the flame. The NO predictions are high by ~50% on the fuel-lean side and this persists to the end of the flame. Predictions show faster consumption of NO on the fuel-rich side and it is under-predicted there by ~20%.

It seems that the chemical mechanism used for methane oxidation is accurate on the fuel-lean side but is inadequate on the fuel-rich side. The NO part of the GRI mechanism gives over-predictions here, opposite to that found in laminar diffusion flames. It is possible that a higher order closure for the conditional reaction rate terms is needed.

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