FIRST-ORDER CONDITIONAL MOMENT CLOSURE MODELLING OF TURBULENT JET DIFFUSION FLAMES OF METHANE

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ABSTRACT

A first-order conditional moment closure (CMC) model, together with fluid flow predictions based on both k-ε and second-moment turbulence closures, is used in the prediction of turbulent jet diffusion flames of methane, with chemical kinetic information supplied from the GRI-Mech 2.11 scheme. To provide an assessment of the ability these methods to predict a range of flames, comparisons are made with experimental data on three piloted and two un-piloted flames. It is concluded that first-order CMC modelling is capable of yielding reliable predictions for flames with little or no extinction effects, with the only exception being NO that is over predicted at most locations within the flames. Results derived using the two turbulence closures are in general in close accord, with calculations for flames that exhibit extinction effects demonstrating a gradual deterioration of results with increasing Reynolds number, pointing to the requirement for second-order CMC modelling as such effects become significant. Anomalies are observed in regard to predictions of NO which is significantly over predicted for the piloted flames, but only slightly over predicted for the un-piloted flames. These results demonstrate a requirement not only for second-order CMC modelling, but also a need to examine the application of different kinetic schemes, including a detailed investigation of NO chemistry kinetic mechanisms.

INTRODUCTION

Mathematical modelling of turbulent combustion processes is used to assist the design and analysis of practical combustion devices for efficiency improvement and emission reduction. Currently, the requirement to accurately predict pollutant emissions has increased the need for linking turbulent flow calculations and finite-rate chemistry effects. Several methodologies are available for modelling such interactions, including the transported probability density function (PDF) approach and the conditional moment closure (CMC) method. The stochastic PDF method (Pope, 1985) provides a rigorous approach to the inclusion of finite-rate chemistry effects, although significant computing resources are required in using this technique. In contrast, the deterministic CMC approach (Klimenko and Bilger, 1999) provides a more economical method that can be readily integrated within computations of complex practical devices.

The CMC method has been shown to be a promising technique for predicting a wide range of practical problems. These include premixed and non-premixed combustion, relatively slow chemistry effects, and ignition and extinction phenomena. For the turbulent diffusion flames that are the subject of the present work, first-order CMC models have been shown to successfully predict parabolic jet flames of H_2/He (Barlow et al., 1999), CO/H_2/N_2 (Smith, 1994), CO_2/H_2/N_2 (Roomina, 1998), and CH_3OH (Roomina and Bilger, 1999) when no extinction effects are present. The method has also been applied successfully to elliptic, bluff-body stabilised flames (Kim et al., 2000). In addition, second-order corrections of the conditional mean reaction rate terms have been implemented to improve the prediction of flame extinction effects and emissions for jet flames of H_2/He (Kronenburg et al., 1998).

This paper describes the use of a first-order CMC model in calculations of CH_4 jet diffusion flames performed using a detailed chemical kinetic scheme, and with both k-ε and second-moment turbulence closures. Attention is focussed on two simple jet flames of CH_4/H_2/N_2 studied by Meier et al. (2000), and three piloted CH_4/air flames measured experimentally by Barlow and Frank (1998). Roomina and Bilger (2001) have previously used the CMC method to predict one of the piloted flames using a k-ε turbulence model and a variety of kinetic mechanisms. In contrast to this earlier CMC study that focussed on a single flame, the present work aims to provide a comprehensive assessment of the first-order CMC model's capabilities by considering a range of flames.

MATHEMATICAL MODEL

Solutions of the two-dimensional, axisymmetric forms of the density-weighted flow equations, supplemented with k-ε (Jones and Launder, 1972) and second-moment (Jones and

Musonge, 1988) turbulence closures, were used to obtain flow and mixing field predictions. Closure of the mean density term was achieved using a prescribed β-PDF, with instantaneous values of density derived from adiabatic, equilibrium calculations based on GRI-Mech 2.11 (Bowman et al., 1996). Standard constants were employed in both the turbulence models, apart from $C_{\varepsilon 1}$ that was modified between the standard and accepted value of 1.6 to improve spreading rate predictions. Mean and variance equations for mixture fraction were solved in conjunction with the k-ε model, with an improved version (Fairweather et al., 1992) of the original scalar flux model implemented in the case of the second-moment closure. Solution of the transport equations was achieved using a modified version of the GENMIX code (Spalding, 1977) that employed expanding finite-difference meshes within a stream function formulation, and in all cases grid-independent solutions were established using resolutions in excess of one million nodes.

A first-order, parabolic CMC model was implemented, based on the set of equations that describe the production and transport of conditionally averaged species mass fractions and enthalpy (Klimenko and Bilger, 1999). For the jet flames modelled, the simplifying assumptions of negligible macro-transport by molecular diffusion and turbulent flux contributions were invoked (Klimenko and Bilger, 1999). Since jet flames display a large degree of radial independence of conditional statistics (Bilger, 1993), radial terms were evaluated using cross-stream averaged velocity and scalar dissipation values, as defined by Klimenko (1990). The conditional axial velocity appearing in the descriptive equations was modelled as a PDF-weighted, cross-stream-averaged value, with the approach of Girimaji (1992) used to represent the conditional scalar dissipation. Non-linear conditional source terms were approximated as for first-order closure, assuming the fluctuations of production rate around the mean to be negligible. Mean values were obtained using the CHEMKIN package (Kee et al., 1996) employing the GRI-Mech 2.11 scheme that consists of 277 reactions involving 49 species. Predictions were also obtained using GRI-Mech 3.0 (Smith et al., 2001) that uses 325 reactions involving 53 species. The conditional enthalpy equation was solved with the source term taken to be the conditional radiation heat loss, modelled using the optically thin assumption as outlined by Marracino and Lentini (1997).

Flow and mixing field information from turbulent flow calculations employing a reacting flow density were passed to the CMC model, where the set of species mass fraction equations plus the enthalpy equation were solved in mixture fraction space. Comparison between densities obtained from the CMC solution and prescribed equilibrium values showed little variation at the majority of locations examined in the flames considered. Coupling of the flow field and CMC calculations was therefore not performed for the majority of the calculations reported. Solution of the CMC equations in real space was achieved using a fractional step method, implemented using the stiff ODE solver VODE (Brown et al., 1989) which applies a backward differentiation formula approach to solution of the non-linear equation set. Second-order differential sample space terms were determined using a central differencing approximation. In all cases, the spatial resolution was in excess of $3x10^3$ nodes.

Meier et al. (2000) considered two jet diffusion flames of $CH_4/H_2/N_2$, designated Flames A and B. Fuel issued from a stainless steel tube with an inner diameter of 8 mm at velocities of 42.2 and 63.2 m s^{-1}, with a co-flow velocity of 0.3 m s^{-1} being used in both cases. The higher Reynolds number flame was close to the blow-off condition, and considered to exhibit some localised extinction. Barlow and Frank (1998) considered three piloted turbulent CH_4/air diffusion flames, designated Flames C, D and E. The burner geometry consisted of an axisymmetric fuel jet, of diameter 7.2 mm, surrounded by a pilot annulus. Fuel issued from the central nozzle at 29.7, 49.6 and 74.4 m s^{-1}, with an ambient air co-flow of 0.9 m s^{-1} used in all cases. Flames C and D were considered to exhibit little local extinction and Flame E moderate extinction effects. In modelling these flames inlet boundary conditions were prescribed from experimental data. Computations of Flames A and B assumed fuel to issue from a straight pipe into a co-flow. For Flames C, D and E, testing of various initial conditions for the jet, pilot and co-flow streams demonstrated that closest agreement with downstream velocity and mixture fraction data was obtained when flat distributions for the jet and co-flow streams were used. Results derived using flat profiles are therefore used in the following section.

RESULTS AND DISCUSSION

Space restrictions preclude a detailed consideration of the accuracy of the velocity and mixing field predictions. However, in the case of Flames A and B, predictions of radial profiles of mean mixture fraction and its fluctuations, mean axial velocity and its fluctuations, and shear stress were in good agreement with data. Little difference was apparent between results based on the two turbulence modelling approaches, although the second-moment closure provided more accurate results for fluctuating axial velocities. Results for Flames C to E, which included information on radial velocity fluctuations, were generally in line with those for Flames A and B, although for these flames the superiority of the second moment closure was not only demonstrated for fluctuating axial velocities, but also for fluctuating mixture fractions and radial velocities. Over all five flames, the Reynolds stress results were in closer agreement with data than the alternative k-ε predictions, with all the predictions demonstrating a similar level of agreement with data to that found by Roomina and Bilger (2001) in their study of Flame D.

Measured and predicted conditional species mass fractions and temperatures at x/d (centre line distance/pipe diameter) = 20 in Flames A and B are compared in Figures 1 and 2. Results for Flame A demonstrate good agreement with data, although H_2O and CO tend to be slightly under predicted, and CO_2 and OH over predicted, for fuel-rich conditions. There is also a slight over prediction of temperatures at mixture fraction values between 0.6 and 0.8. Agreement for NO is, however, good, despite a slight over prediction around stoichiometric and in fuel-rich regions. By x/d = 60 predictions (not shown) for major species and temperature were in line with observations since the probability of encountering fuel-rich regions is low, although the under and over prediction, respectively, of CO and OH remained. Results for NO were also seen to over predict more

significantly under fuel-lean conditions. Results derived from the two turbulence models showed little difference at all measurement locations, in line with velocity field results. Results for Flame B (Figure 2) generally confirm these conclusions, although temperature predictions were in good agreement with data at all locations examined in this flame. However, results for NO now over predict data at all downstream distances, and more significantly than for Flame A. Results derived for both flames using GRI-Mech 3.0 confirmed the trends noted above, with very close agreement with predictions made on the basis of GRI-Mech 2.11, apart from NO, shown in Figure 3, where the absolute level of this species is seen to be approximately doubled at all mixture fractions, leading to significant over prediction of data.

Equivalent results at x/d = 30 in Flames C to E are shown in Figures 4 to 6. Compared to the earlier comparisons for Flames A and B, results for Flame C (Figure 4) are less satisfactory and now show an over prediction of temperature at all fuel-rich mixture fractions. Also, in contrast to results for Flames A and B, CO, H_2 and H_2O are now over predicted, and CO_2 under predicted, in fuel-rich regions, with CH_4 and O_2 also significantly under predicted in these regions. The over prediction of temperatures, and under prediction of CH_4 and O_2, under fuel-rich conditions should lead to an over prediction of CO_2 and H_2O. This does occur for the latter species, but CO_2 is slightly under predicted due to the overestimation of CO levels. NO levels are also over predicted more significantly than in the previous flames, with deviation from experiment being greatest under fuel-lean and near-stoichiometric conditions. Further downstream at x/d = 60 predictions (not shown) came more in line with observations, again because the probability of encountering fuel-rich regions is very low. The only exception was NO where a significant over prediction remained. Similar results were obtained at all other measurements stations examined within this flame, with results obtained using the two turbulence closures being in close agreement at all locations, although CO and H_2 predictions obtained with the second-moment closure were marginally superior.

With increasing Reynolds number (Figure 5 and 6), and increasing extinction effects, the trends between measured and predicted flame characteristics observed in Flame C remain, although the deviation between the two sets of results increases. The only exceptions are CO_2, where an under prediction at low Re turns into an over prediction at high Re, and NO, which is in marginally better agreement with data for Flame D. The accuracy of the predictions does, however, increase with downstream distance in each flame, for the reasons noted above. For Flame E, which is considered to exhibit moderate extinction effects, significant variations exist between predicted and observed results. All these results, including those for Flames A and B, point to a requirement for the use of second-order CMC modelling as extinction effects become more significant. Results derived for all these flames using GRI-Mech 3.0 confirmed the trends noted above, apart from NO, shown in Figure 7, where absolute levels were again approximately doubled at all mixture fractions leading to a significant over prediction of data.

The results for Flame D are in good agreement with those obtained by Roomina and Bilger (2001), although differences do occur for NO. In particular, predicted NO levels obtained in the present work over estimate data in fuel-lean and near-stoichiometric regions, but come in line with data under very fuel-rich conditions. In contrast, the predictions of Roomina and Bilger (2001), whilst qualitatively similar to those of Figure 4, under predict NO levels under very fuel-rich conditions.

With the exception of NO, agreement of predictions of the first-order CMC model and data is in general adequate for Flames A, B, C and D, although results for Flame E are not. The increasing importance of flame extinction effects, exhibited by an increasing divergence from observations as Reynolds numbers are increased from Flame A to B, and from Flame C to D to E, illustrates that first-order closure of the conditional mean reaction rates becomes inaccurate with significant conditional fluctuations of species concentrations and temperature. Allowance for the variances and co-variances of the fluctuations to improve the conditional mean reaction rates within a second-order CMC model is therefore required for such flames.

Uncertainties also remain in regard to predictions of NO. Whilst results for Flames A and B are in reasonable agreement with measurements, those for Flames C to E are not. The present work was performed on the basis of GRI-Mech 2.11 and 3.0 and, whilst obtaining close agreement between the results of these two schemes for the majority of species and temperature, the NO results exhibit a significant discrepancy. These findings are in line with those of Roomina and Bilger (2001) who used a variety of kinetic schemes to predict Flame D, including GRI-Mech 2.11. As noted above, the GRI-Mech 2.11 based results obtained by these authors were similar to those of the present study, although some differences did occur under fuel-rich conditions. In particular, good agreement between predictions of temperature and reactive scalars was obtained in fuel-lean regions, with an over prediction of NO in fuel-lean regions and an under prediction when fuel-rich. Roomina and Bilger (2001) attributed errors on the fuel-lean side to either the use of a first-order approximation for closure on conditional reaction rate terms within the CMC model employed, or the need for further work to establish whether the rate constant for the N_2O pathway to NO formation, which is important in fuel-lean regions, is too high. On the fuel-rich side, the comparison of different mechanisms revealed the importance of C_2 chemistry, although the overall adequacy of current mechanisms in fuel-rich regions was questioned, and the need for further model improvement and wider validation against a range of non-premixed flames identified. The present work largely confirms these findings in regard of Flames C to E, although reasonable agreement for NO levels is obtained in fuel-rich regions of Flame C, with no under prediction of NO levels being seen in any of the three flames. Predictions for Flames A and B show much closer agreement with NO data at all stoichiometries, although over prediction in both fuel-lean and fuel-rich zones is generally the case. Results for NO are therefore conflicting, and demonstrate a need to explore second-order CMC modelling approaches since the inclusion of such terms has been found (Kronenburg et al., 1998) to significantly reduce NO levels over all stochiometries. In addition, however, there is clearly a requirement to further investigate the application of different

kinetic schemes in the modelling of turbulent non-premixed flames, including detailed investigations of the mechanisms and rates employed for NO chemistry.

Lastly, although not shown, comparisons were made between data and predictions obtained from fully coupled flow field and CMC calculations, both in conditional and real space. As noted earlier, the conditional space predictions varied negligibly from those obtained without coupling. Real space predictions, although less informative about the level of agreement with data than the conditional averaged statistics considered so far, do demonstrate the accuracy that may be expected in practical applications. By way of illustration, for Flame A the trends observed in the conditional results shown above were reproduced in the coupled predictions, although predictions of temperature, CO_2 and H_2O tended to be more in line with data, whilst the under estimation of CO, and over estimation NO and OH, remained. Oxygen was also under predicted at all radial positions. Overall, however, the predictions showed good agreement with data, and at virtually all locations results obtained using the second-moment turbulence closure were superior to k-ε based results, despite little difference being apparent between predictions based on these two models in conditional space.

CONCLUSIONS

The work described in this paper provides an assessment of the ability of first-order CMC methods to predict a range of turbulent, non-premixed methane flames. First-order CMC modelling is found to be capable of yielding reliable predictions for flames that have little or no extinction effects, with the exception of NO levels. There is a general deterioration of results from Flames A to B, and from C to E, pointing to the requirement for second-order CMC modelling as local extinction effects become significant. There is little difference between results derived on the basis of k-ε and Reynolds stress turbulence models for the majority of flow parameters considered, in conditional space at least. Real space predictions did, however, demonstrate the superiority of results derived on the basis of the second-moment turbulence closure. Anomalies remain in regard to predictions of NO that is over predicted at most locations and stoichiometries on all the flames considered. Results for Flames C to E demonstrate significant over prediction, whilst those for Flames A and B are in closer accord with data. These results demonstrate a requirement not only to explore second-order CMC modelling approaches, but also to examine the application of different kinetic schemes, including a detailed investigation of the mechanisms and rates employed for NO chemistry.

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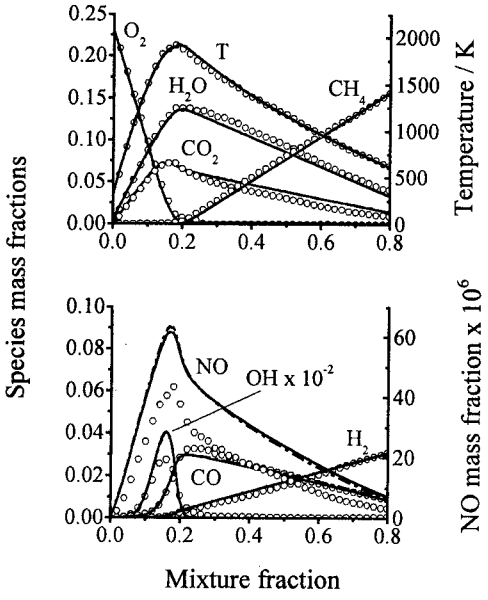

Figure 2. Conditional species mass fractions and temperatures at x/d = 20 in Flame B (key as Figure 1).

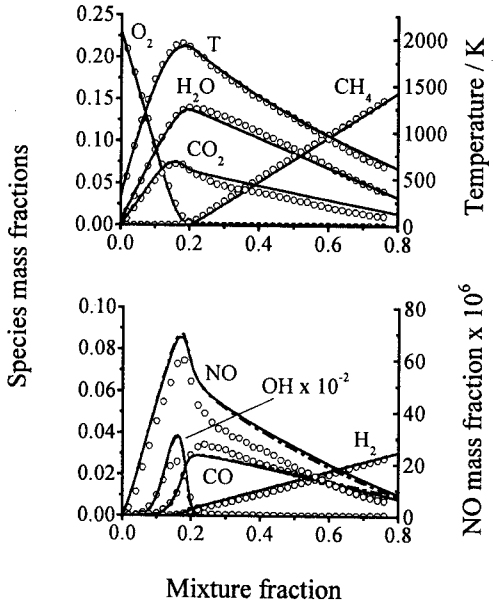

Figure 1. Conditional species mass fractions and temperatures at x/d = 20 in Flame A (circles - measured, solid line - predicted Re stress, dashed line - predicted k-ε).

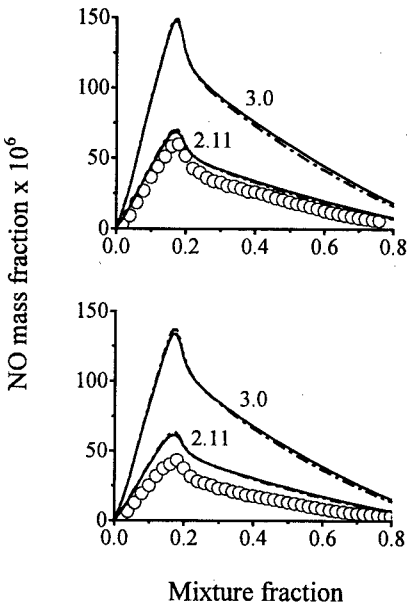

Figure 3. Conditional mass fractions of NO at x/d = 20 in Flames A and B obtained using GRI-Mech 2.11 and 3.0 (key as Figure 1, top Flame A, bottom Flame B).

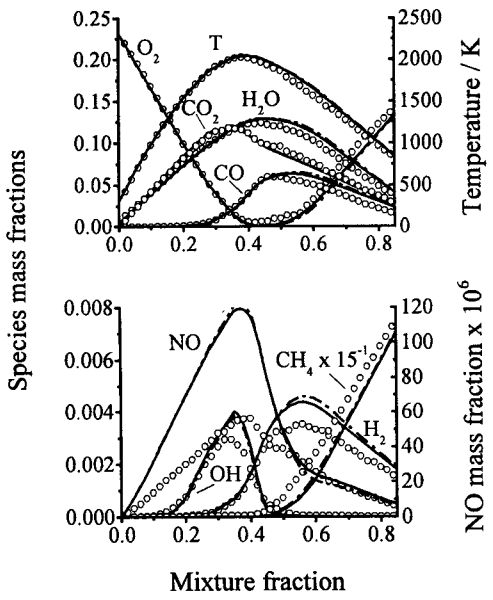

Figure 4. Conditional species mass fractions and temperatures at x/d = 30 in Flame C (key as Figure 1).

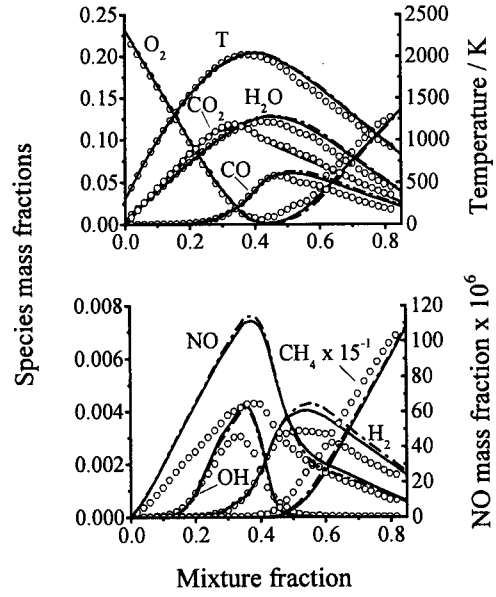

Figure 5. Conditional species mass fractions and temperatures at x/d = 30 in Flame D (key as Figure 1).

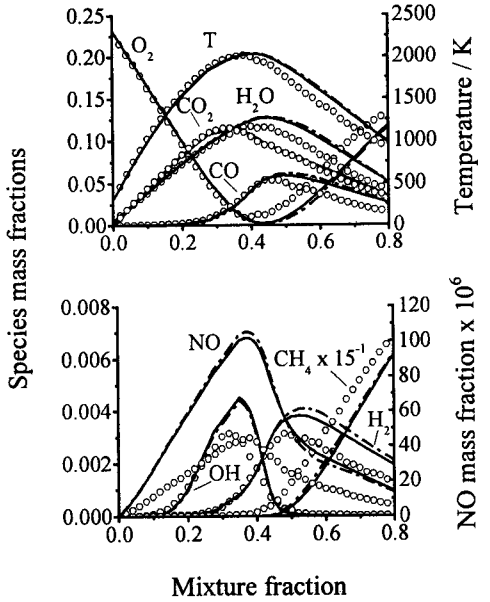

Figure 6. Conditional species mass fractions and temperatures at x/d = 30 in Flame E (key as Figure 1).

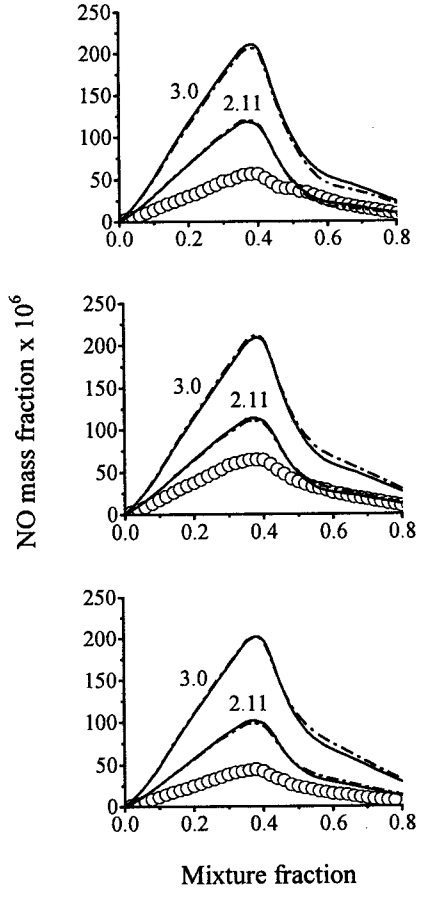

Figure 7. Conditional mass fractions of NO at x/d = 30 in Flames C to E obtained using GRI-Mech 2.11 and 3.0 (key as Figure 1, top Flame C, bottom Flame E).