Transported PDF Modeling of Nonpremixed Turbulent CO/H$_2$/N$_2$ Jet Flames

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TRANSPORTED PDF MODELING OF NONPREMIXED TURBULENT CO/H₂/N₂ JET FLAMES

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Turbulent CO/H₂/N₂ (“syngas”) flames are simulated using a transported composition probability density function (PDF) method. A consistent hybrid Lagrangian particle Eulerian mesh algorithm is used to solve the modeled PDF transport equation. The model includes standard k-ε turbulence, gradient transport for scalars, and Euclidean minimum spanning tree (EMST) mixing. Sensitivities of model results to variations in the turbulence model, the treatment of radiation heat transfer, the choice of chemical mechanism, and the PDF mixing model are explored. A baseline model reproduces the measured mean and rms temperature, major species, and minor species profiles reasonably well, and captures the scaling that is observed in the experiments. Both our results and the literature suggest that further improvements can be realized with adjustments in the turbulence model, the radiation heat transfer model, and the chemical mechanism. Although radiation effects are relatively small in these flames, consideration of radiation is important for accurate NO prediction. Chemical mechanisms that have been developed specifically for fuels with high concentrations of CO and H₂ perform better than a methane mechanism that was not designed for this purpose. It is important to account explicitly for turbulence-chemistry interactions, although the details of the mixing model do not make a large difference in the results, within reasonable limits.

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Keywords: Nonpremixed turbulent flames; Probability density function method; Syngas flames

INTRODUCTION

The replacement of air with pure oxygen in coal-fired systems (oxy-coal combustion) is one option that is being considered for next-generation stationary power generation. The N₂-lean exhaust stream facilitates the separation of CO₂ from
the other combustion products, mainly $\text{H}_2\text{O}$. This facilitates sequestration, thus reducing the emission of $\text{CO}_2$ into the atmosphere. A reduction in NOx (Buhre et al., 2005) and other pollutants has also been seen in pilot-scale studies.

The thermochemical environment differs substantially from that of conventional air-based combustion because of higher concentrations of species including $\text{O}_2$, $\text{CO}_2$, and $\text{H}_2\text{O}$. This differing environment poses a challenge for the computational models and related experimental data that have focused on air combustion. Also, gaseous radiation heat transfer is expected to be more prominent due to the higher concentration of $\text{H}_2\text{O}$ and $\text{CO}_2$. This should also amplify the importance of accurate modeling of the turbulence–radiation interactions, which often have been neglected in CFD calculations.

Here turbulent $\text{CO}/\text{H}_2/\text{N}_2$ ("syngas") flames have been simulated, as a first step toward modeling oxy-coal combustion. The product composition is similar to that of the flue gases in coal- or biomass-fueled combustion systems (Zahirovic et al., 2006), without the complications of solid fuels. The gas-phase chemistry is relatively simple, while finite-rate chemistry and turbulence–chemistry interactions are important because of the slow and indirect oxidation of CO to $\text{CO}_2$ (Abián et al., 2011; Barlow et al., 2000; Li et al., 2007; Zsely et al., 2005). The flame configurations are those targeted by the International Workshop on Measurement and Computation of Turbulent Nonpremixed Flames (TNF Workshop) (Barlow, 2011), for which detailed measurements are available. Compared to other TNF Workshop flames (the piloted methane/air nonpremixed turbulent jet flames, in particular), relatively few modeling studies have been reported for the TNF syngas flames. Two difficulties from a modeling perspective are flame stabilization and potential differential diffusion; these are discussed in subsequent sections.

Transported probability density function (PDF) methods are one of the best available approaches for dealing with complex nonlinear interactions in turbulent combustion (Pope, 1985). PDF methods have been tested and applied to configurations from canonical laboratory flames to practical devices including gas-turbine combustors and reciprocating-piston engines (Haworth, 2010). For exploration of new combustion regimes such as oxyfuel, PDF methods with relatively detailed chemistry offer added advantages over simpler models that have more flow- and chemistry-specific tuning parameters. To date, no transported PDF modeling studies have been reported for the TNF workshop syngas flames. Here, a consistent hybrid Lagrangian particle/Eulerian mesh method is used to solve a modeled transport equation for the one-point, one-time joint PDF of species mass fractions and mixture specific enthalpy. A comprehensive set of physical models and numerical algorithms is integrated and validated, including a spectral photon Monte Carlo method for participating-medium radiation. Sensitivities of computed results to variations in the gas-phase chemical mechanism, the radiation heat transfer model, and the PDF mixing model are explored.

The remainder of the article is organized as follows. In the following section, the target flames are introduced and findings from earlier modeling studies are summarized. Then, the physical models and numerical methods are described. Comparisons with experimental measurements, including sensitivities to model variations, are reported next. In the final section, key findings are summarized and next steps are outlined.
TNF WORKSHOP SYNGAS FLAMES

Two turbulent jet flames of 40% CO, 30% H₂, 30% N₂ by volume issuing into coflowing air are simulated (Table 1). Flame A has a smaller fuel-jet diameter and higher velocity compared to flame B, such that the fuel-jet-based Reynolds numbers of the two flames are the same: \( Re_f = 16500 \). Available experimental data include radial profiles of mean and rms axial velocity, temperature, and species mass fractions at several distances downstream of the nozzle. Details can be found in Barlow et al. (2000) and Barlow (2011). Earlier modeling studies for these flames have been reported in Kim et al. (2001), Cuoci et al. (2007), Frassoldati et al. (2007), Zahirovic et al. (2001, 2006), and Hewson and Kerstein (2001).

Kim et al. (2001) used a Reynolds-averaged formulation with a \( k-\varepsilon \) turbulence model. A steady flamelet model and an unsteady flamelet model based on a Lagrangian approach were used to account for turbulence-chemistry interactions. Computed profiles of mean velocity and major species concentrations for flames A and B were shown to coincide when plotted as functions of normalized (by the fuel-jet-nozzle diameter) spatial coordinates, consistent with the experimental measurements. An optically thin model was used for radiation. Radiation effects were shown to be small in both flames, with calculated radiant fractions of approximately 7% and 17% for flames A and B, respectively, which are higher than the experimental values (3.4% and 7.1%, respectively).

Cuoci et al. (2007) reported Reynolds-averaged simulations using a \( k-\varepsilon \) turbulence model and three different turbulence-chemistry interaction models. Simple chemical mechanisms were used first in coupled calculations to compute the temperature, major species, and flow fields. Then a detailed chemical mechanism (Frassoldati et al., 2007) was used in a kinetic post-processing step to predict the formation of pollutants including NOx. The eddy-dissipation-concept (EDC) and steady laminar flamelet (SLF) models showed better agreement with experiment compared to a simpler eddy dissipation (ED) model. Discrepancies between computed and measured CO were found, especially far downstream of the nozzle. Improved NOx predictions were found when the temperature fluctuations were accounted for explicitly in the post-processing. The computed peak mean temperature dropped by 30–40 K with a discrete-ordinates radiation model, compared to neglecting radiation.

Similar simulations were reported by Zahirovic (Zahirovic et al., 2001, 2006), using a realizable \( k-\varepsilon \) turbulence model with EDC, SLF and ED turbulence-chemistry

<table>
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<th>Table 1 Inlet specifications for syngas flames A and B (Species compositions are mass fractions)</th>
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<td>Stream</td>
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<td>Diameter (mm)</td>
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interaction models. The purpose was to validate the use of these models for biogas combustion. Results from four different chemical mechanisms were compared: a global mechanism, a nine-species augmented reduced mechanism, a 19-species augmented reduced mechanism, and a detailed 32-species, 153-reaction mechanism. All mechanisms except the global mechanism yielded similar results.

Hewson and Kerstein (2001) applied the one-dimensional turbulence (ODT) model to these flames, and studied the evolution of the velocity field as well as the first and second moments of the reacting scalars conditioned on the mixture fraction. Radiation was neglected. Because molecular transport can be resolved with this model, the importance of differential diffusion could be explored. It was reported that differential diffusion is important close to the nozzle (to approximately six fuel-jet diameters downstream), where experimental data are not available. The importance of differential diffusion also was explored in Barlow et al. (2000), by comparing the results from laminar flame calculations with equal diffusivities to those with full differential diffusion. There it was concluded that differential diffusion plays a relatively small role in these flames, especially far downstream of the nozzle. In the context of transported PDF methods, molecular transport is accounted for through a mixing model. Some mixing models have been designed to consider differential diffusion (Fox, 2003; Viswanathan et al., 2011). However, these models are still at an early stage of development, and it would be better to explore differential diffusion in a configuration where the effects are less ambiguous. In the present study, differential diffusion is neglected.

Although no transported PDF modeling studies have been reported for the TNF workshop syngas flames, several early transported PDF modeling studies focused on similar CO/H2/N2 nonpremixed jet flames (Chen et al., 1991; Correa et al., 1985, 1988; Haworth et al., 1988, 1989; Pope and Correa, 1987), using various chemical mechanisms and other models. The early studies were limited by the computational power and chemical mechanisms that were available at the time. Rudimentary parabolic solution algorithms were used, and radiation heat transfer was not considered. In present study, modern elliptic PDF solution algorithms are used with newer chemical mechanisms, and radiation heat transfer is considered.

PHYSICAL MODELS AND NUMERICAL METHODS

A modeled transport equation for the joint PDF of species mass fractions and mixture specific enthalpy is solved using a consistent hybrid Lagrangian particle/Eulerian mesh (finite-volume) method using a Reynolds-averaged formulation. The physical models and numerical methods are introduced in subsequent subsections. Further information can be found in Sections 6 and 7 of Haworth (2010), and in the other cited references.

Hybrid Lagrangian Particle/Finite-Volume PDF Method

In a composition PDF method, models are required for turbulence, turbulent scalar transport, and molecular transport. No further modeling (beyond specification of thermochemical and radiative properties and a chemical reaction mechanism)
is required to account for the effects of turbulent fluctuations in composition and temperature on mean chemical reaction rates or mean radiative emission.

Here turbulence is modeled using a standard two-equation $k-\varepsilon$ model, gradient transport is assumed for turbulent scalar transport, and molecular transport is modeled using either a modified Curl (MC) (Curl, 1963; Nooren et al., 1997) or a Euclidean minimum spanning tree (EMST) (Masri et al., 1996; Subramaniam and Pope, 1998) mixing model. In general, EMST is expected to be the better mixing model, as it takes into account the locality of the mixing in composition space; in particular, EMST has been shown to perform better at higher Damköhler numbers. Differential diffusion is neglected, in all cases. Standard values have been used for all model coefficients, with two exceptions. First, the value of the mixing model coefficient, $C_\phi$ (the ratio of a turbulence hydrodynamic time scale to a turbulence scalar time scale), has been varied to explore its influence on model results. Here the baseline mixing model is EMST with $C_\phi = 1.5$. It should be noted that a somewhat higher value of $C_\phi$, usually has been used in PDF-based modeling studies of turbulent jet flames using the MC mixing model ($C_\phi \approx 2.0$) compared to the EMST model ($C_\phi \approx 1.5$). Second, the value of $C_{s1}$ in the $\varepsilon$ equation has been increased from its standard value of 1.44 to 1.6 (James et al., 2001). In general, a higher value of $C_{s1}$ lowers the computed jet spreading rate. It is conventional practice in $k-\varepsilon$ modeling studies of circular jet flames to increase the value of $C_{s1}$ (Jaishree and Haworth, 2012). The influence of a small variation in $C_{s1}$ on computed mean profiles is demonstrated below.

The underlying finite-volume CFD solver is OpenFOAM 1.5 (OpenFOAM, 2011). A PISO-based, time-implicit segregated solver is used to solve the coupled mean momentum, pressure, energy, and $k-\varepsilon$ equations using second-order spatial discretizations. Species mass fractions and mixture specific enthalpy are computed on the particle side, and local mean scalar values are estimated as appropriately weighted averages over particle values. The principal feedback from the particle side to the finite-volume side of the calculation is through the mean density; this is handled using an “equivalent enthalpy”-based approach (Muradoglu et al., 1999, 2001). Particle/finite-volume coupling and other numerical issues are discussed in detail in Haworth (2010). Solutions are advanced in time starting from (essentially) arbitrary initial conditions until a statistically stationary state is reached. Following the recommendation given in Jaishree and Haworth (2012), the nominal number of particles per finite-volume cell is 40.

**Thermochemical Properties and Chemical Mechanisms**

A 10-species, six-reaction mechanism (including NO) that was developed specifically for CO/H$_2$/N$_2$ mixtures in the proportions of interest here (40%/30%/30%) (Chen, 2011) has been adopted as the baseline for this study. This relatively low-computational-cost mechanism is well suited for performing parametric studies on the influences of variations in the other physical submodels (e.g., radiation and mixing). The influence of the chemical mechanism is also of interest, especially for predictions of NO and other minor species. This has been explored by comparing results obtained using the baseline mechanism with those obtained using two other mechanisms: GRI-Mech 2.11 (Bowman et al., 1995) (which includes NOx) and a recently updated Cl mechanism (Li et al., 2007) that does not include NOx.
In the latter mechanism, new values of rate constants have been recommended for the reactions \( \text{CO} + \text{OH} = \text{CO}_2 + \text{H} \) and \( \text{HCO} + \text{M} = \text{H} + \text{CO} + \text{M} \), which are important in \( \text{CO}/\text{H}_2 \) systems. The model also has a modified heat of formation for the \( \text{OH} \) radical. These changes are expected to have a significant influence on the \( \text{CO} \) prediction and consequently the temperature (Abián et al., 2011; Li et al., 2007; Zsely et al., 2005). They are also expected to have a significant influence on \( \text{NO} \) prediction in situations where prompt \( \text{NO} \) is important (Dryer, 2011). That is not the case in the flames that are simulated here, but it still is of interest to assess this mechanism, as it has been developed specifically for syngas flames.

**Radiative Heat Transfer**

Measured radiant fractions for flames A and B are 3.4% and 7.1%, respectively (Frank et al., 2000). Thus radiation effects are small, and radiation is neglected altogether for the baseline simulations.

Results without radiation are compared to those obtained using a gray, optically thin model. This model considers emission from \( \text{CO}_2 \), \( \text{H}_2\text{O} \), and \( \text{CO} \); the absorption coefficient is prescribed following the approach described in Smith et al. (2011). The syngas flames simulated here have high \( \text{CO}_2 \) concentrations compared to hydrocarbon/air flames; thus absorption in the 4.3\( \mu \text{m} \) \( \text{CO}_2 \) band is expected to be relatively high. While it is not anticipated that spectral radiation and reabsorption influence the global structure of these flames significantly, advanced radiation treatments will be required as we progress toward oxy-coal combustion. Therefore, a full spectral photon Monte Carlo method that has been used in earlier PDF-based modeling studies (Mehta et al., 2010; Wang and Modest, 2006) also has been tested here. This model maintains essentially line-by-line spectral accuracy, and considers emission and absorption by \( \text{CO}_2 \) and \( \text{H}_2\text{O} \).

Results obtained using three different levels of radiation treatment are compared in the results section: No radiation (baseline), gray optically thin radiation, and spectral radiation with reabsorption.

**Flame Stabilization**

A practical issue in simulating these syngas jet flames is flame stabilization. In the laboratory, the flame is anchored to the lip of the fuel-jet tube by a small recirculation zone just downstream of the nozzle wall (thickness 0.88 mm) (Barlow et al., 2000; Cuoci et al., 2007; Giacomazzi et al., 2008); no pilot was required and no lift-off was observed in the experiments. This anchoring mechanism was confirmed by Giacomazzi et al. (2006), who performed high-spatial resolution (10\(-5\) m), large-eddy simulation for a 25 mm by 15 mm area near the nozzle wall.

Resolving the recirculation zone in a full-flame simulation with detailed chemistry would be computationally prohibitive. It is expedient to use a coarser mesh and to introduce an artificial anchoring mechanism, and that is the approach that has been taken in most of the earlier modeling studies. The extent to which artificial stabilization is necessary depends on the chemical mechanism that is used. Cuoci et al. (2007) stabilized the flame by including a short upstream section of the fuel tube in their computational domain; the computational cost was kept down through
the use of small chemical mechanisms, with the use of detailed chemistry limited to a post-processing step. In the simulations of Zahirovic et al. (2006), a small high-temperature zone was introduced at a location just downstream of the nozzle to act as an artificial pilot.

For the computational meshes and chemical mechanisms that are used in this study, the simulated flames blow out in the absence of an artificial anchoring mechanism. Therefore, a small zone near the nozzle is designated as the ignition zone. In the ignition zone, a local equilibrium calculation is performed for any computational particle whose mixture fraction is within 2% of the stoichiometric value. This corresponds to approximately $6 \times 10^{-6}$% (by mass) of the total fluid in the computational domain. A sensitivity study has been done to minimize the extent of the ignition zone, and to confirm that it has negligible influence on computed mean and rms temperature and major species profiles. On the other hand, NO chemistry (dominated by thermal NO here) is slow, and local equilibrium may lead to unrealistically high local NO levels. To mitigate this, particle NO values in the ignition zone are set to zero.

Computational Mesh, Initial and Boundary Conditions

The computational domain is a 10-degree wedge with a single finite-volume cell in the azimuthal direction, corresponding to axisymmetric simulations. The inlet is at the plane of the fuel-jet nozzle exit, and the domain extends 80d in the axial direction and 15d in the radial direction, where $d$ is the fuel-jet diameter. A nonuniform mesh of 2700 finite-volume cells is used, with finer resolution close to the fuel nozzle and mixing zone.

In earlier PDF-based modeling studies for nonpremixed turbulent jet flames, little difference has been found between results obtained using a top-hat inlet mean velocity profile and measured inlet mean velocity profiles (where available). Here, top-hat profiles are specified for mean velocity, composition, and temperature at the inlet using the values given in Table 1. Inlet values of turbulence kinetic energy ($k$) are specified to be the experimental values. Inlet values of dissipation rate of turbulence kinetic energy ($\varepsilon$) are specified to correspond to a turbulence integral length scale of $l_T = 0.03d$; here $\varepsilon = C_{\mu}^{0.75}k^{1.5}/l_T$, where $C_{\mu} = 0.09$ is a standard $k-\varepsilon$ model constant. At the outlet, a fixed pressure of one atmosphere is specified and zero-gradient conditions are used for all other quantities. Zero gradient conditions are applied at the outer radial boundary, and symmetry conditions are applied on the azimuthal faces.

Initial conditions in the computational domain correspond to ambient air. The simulations are advanced in time with a computational time step of 5 $\mu$s for flame A and 10 $\mu$s for flame B (corresponding to a maximum material Courant number of approximately 0.5), until a statistically stationary state is reached (approximately 2s, or 200 flow-through times based on the fuel-jet mean velocity). Results then are time-averaged for approximately 0.5 s (50 flow-through times) to reduce statistical noise in the reported mean and rms profiles.

Computational Acceleration

The simulations have been run on small multiprocessor systems using up to eight cores. Simple domain decompositions have been performed in the axial direction, and in some cases an additional level of parallelization has been implemented
for the particle chemistry calculations (Haworth, 2010; Jaishree and Haworth, 2012). In most cases, in situ adaptive tabulation (ISAT) (Lu and Pope, 2009) has been used to accelerate the computation of the chemical source terms. Verification that results obtained using ISAT are essentially the same as those obtained by direct integration of the chemical source terms is provided in the following section.

RESULTS AND DISCUSSION

The calculation of chemical source terms dominates the computational effort, and it is desirable to reduce this cost using ISAT. The accuracy and efficacy of using ISAT is verified first. Then results obtained using the baseline models (with ISAT) are presented. This is followed by discussions of the influences of variations in four aspects of the physical modeling: the turbulence model ($C_{\mu}$), radiation, the chemical mechanism, and the PDF mixing model. All mean and rms profiles correspond to mass- (Favre-) averaged quantities. The figures that are included in the article represent a subset of the available results. A more complete set of figures is provided in the Supplementary Material that is available online.

ISAT

A key ISAT parameter is the global error tolerance, $\varepsilon_{\text{ISAT}}$. Smaller values of $\varepsilon_{\text{ISAT}}$ correspond to higher accuracy and higher computational cost. In most PDF modeling studies, values between $10^{-4}$ and $10^{-3}$ have been used, and $\varepsilon_{\text{ISAT}}=10^{-3}$ has been shown to be satisfactory for most purposes (Jaishree and Haworth, 2012; James et al., 2001). Here, results obtained using ISAT with $\varepsilon_{\text{ISAT}}=10^{-4}$ and with $\varepsilon_{\text{ISAT}}=10^{-3}$ have been compared with results obtained using direct integration of the chemical source terms. The largest differences are in minor species profiles. An example (mean and rms radial OH profiles at $x/d=40$ for flame A) is shown in Figure 1. With $\varepsilon_{\text{ISAT}}=10^{-3}$, mean temperature and major species profiles are within approximately 5%, and rms temperature and minor species profiles are within approximately 10% of those obtained with direct integration. On a single processor, the overall simulation time for ISAT with $\varepsilon_{\text{ISAT}}=10^{-3}$ is approximately 3% of that for direct

![Figure 1](https://example.com/figure1.png)

**Figure 1** Comparisons between direct integration and ISAT with different tolerances at $x/d=40$. (a) Mean temperature; (b) mean OH gas fraction. (Figure is provided in color online.)
integration. With $a_{\text{ISAT}} = 10^{-4}$, mean temperature and major species profiles are within approximately 2% of those obtained with direct integration, while the differences in rms temperature and minor species profiles remain approximately 10%. The overall simulation time for ISAT with $a_{\text{ISAT}} = 10^{-4}$ is approximately 5% of that for direct integration. All subsequent results have been obtained using ISAT with $a_{\text{ISAT}} = 10^{-3}$.

**Baseline Model Results**

Results for the baseline models ($C_{e1} = 1.6$, no radiation, 10-species chemistry, EMST mixing with $C_{\phi} = 1.5$) are presented next. The predicted flame length based on the mean stoichiometric mixture fraction value ($\bar{\xi}/0.295$) is 49.5 d and 50 d for flames A and B, respectively. This is slightly higher than the measured value of 47 d, and can be improved with a small decrease in $C_{e1}$ (discussed below).

Comparisons of computed and measured radial profiles of mean and rms temperature, major species and minor species at $x/d = 20$ and at $x/d = 40$ for flames A and B are shown in Figures 2 and 3. As can be seen in the figures, scaling by the jet diameter essentially collapses the profiles for the temperature and major species (CO$_2$, H$_2$O, CO), consistent with the experimental findings. Minor species profiles (OH and NO) do not scale simply with nozzle diameter, because of their stronger dependence on local scalar dissipation rate and convective residence time, and the model captures these trends as well (e.g., higher NO for flame B compared to flame A). For all quantities, the level of quantitative agreement between model and experiments is at least as good as any that has been reported in the literature to date, if not better. It is particularly noteworthy that the computed fluctuation levels are in reasonably good agreement with the experiments for all quantities.

The principal discrepancies between model and experiment are in the mean H$_2$O (underpredicted close to the centerline at the more upstream location), the mean CO (overpredicted, especially at the more downstream location), and mean NO. The discrepancy in H$_2$O might be a consequence of neglecting differential diffusion. As discussed earlier, there is evidence that differential species diffusion plays a role in these flames on the fuel-rich side, close to the nozzle (Barlow et al., 2000; Hewson and Kerstein, 2001), because of the high diffusivity of H$_2$. The computed mean CO is very sensitive to the jet spreading rate, as will be shown in the next subsection. The experimental uncertainty for CO is also high (10%) compared to other major species (e.g., 3% for H$_2$). Predicted NO levels depend strongly on the chemical mechanism and the local temperature (hence, radiation). At upstream locations, NO levels may also be influenced by the treatment that has been used to anchor the flame, as discussed earlier. The reported experimental uncertainty for NO is 10% to 15%. The influences of radiation and the chemical mechanism are discussed below.

**Effects of $C_{e1}$**

It is customary practice in $k$-$\varepsilon$ modeling studies of turbulent jet flames to treat $C_{e1}$ as an adjustable parameter that can be tuned to control the spreading rate. The effects of reducing $C_{e1}$ from 1.60 to 1.56 are shown in Figure 4. Reducing $C_{e1}$ increases the spread rate, and shortens the flame. The effect on the downstream
computed mean CO profile is especially pronounced. The relatively high sensitivity of CO might be a consequence of the relatively slow CO chemistry; its chemical time scale is of the same order as the flow time scale (Barlow et al., 2000).

**Effects of Radiation**

As discussed earlier, radiation is relatively weak in these flames. Nevertheless, its effects are discernable. They are most evident at downstream locations for species that are particularly sensitive to small variations in temperature: CO and NO. Thermal NO is dominant for these flames (Frank et al., 2000).
Figure 3 Computed and measured mean and RMS radial profiles of flames A and B, scaled on nozzle diameter at $x/d = 40$. (a) Mean temperature; (b) rms temperature; (c) mean NO mass fraction; (d) rms NO mass fraction; (e) mean CO mass fraction; (f) rms CO mass fraction; (g) mean OH mass fraction; (h) rms OH mass fraction; (i) mean CO$_2$ mass fraction; (j) mean H$_2$O mass fraction. (Figure is provided in color online.)

Figure 5 shows computed mean temperature, CO, and NO profiles at $x/d = 40$ for flames A and B with three radiation models. As expected, the computed mean temperature is highest when radiation is neglected altogether and is lowest when a gray, optically thin model is used; results from the spectral model with reabsorption are between these two extremes. The maximum drop in the computed mean temperature from the no-radiation model to the optically thin model is approximately 40 K for flame A and 100 K for flame B. Lower temperatures result in lower CO and NO, and bring the computed mean CO and NO profiles into closer agreement with experiment.
Figure 4 Comparisons of results obtained using two different values of $C_{el}$ ($C_{el} = 1.6$ and $C_{el} = 1.56$) at $x/d = 40$ for flame B. (a) Mean axial velocity; (b) mean mixture fraction; (c) mean temperature; (d) mean CO; (e) mean CO$_2$; (f) mean OH. (Figure is provided in color online.)

Figure 5 Comparisons of results obtained using three radiation models at $x/d = 40$. (a) Mean CO, flame A; (b) mean NO, flame A; (c) mean CO, flame B; (d) mean NO, flame B. (Figure is provided in color online.)
**Effects of Chemical Mechanism**

Mean and rms temperature, CO₂, H₂O, and OH profiles are similar for the three mechanisms that have been tested. The effects of changes in the chemical mechanism are most evident in computed CO₂, H₂, and NO levels; these are shown at \( x/d = 40 \) in Figures 6 and 7 for flame A. The Cl mechanism does not include NO. Large differences can be seen in computed NO levels between GRI-Mech 2.11 and the 10-species mechanism, in particular. GRI-Mech 2.11 overpredicts the CO₂, H₂, and NO levels for both flames, compared to the two mechanisms that have been developed specifically for syngas.

As pointed out by Frank et al. (2000), only thermal NO should exist in the syngas flames. The additional NO paths in the GRI-2.11 mechanism could lead to the overprediction of NO levels. To assess this assumption, all NO pathways have been removed from the GRI-2.11 mechanism, except the three-step extended Zeldovich mechanism. Figure 7 shows that NO prediction is greatly improved when nonthermal NO is removed, and the results are nearly identical to those obtained using the 10-species mechanism. The improvement seen here supports the conclusion that only thermal NO exists in the syngas flames.

**Effects of the Mixing Model**

In a PDF method, the mixing model plays a central role, especially in situations where finite-rate chemistry and turbulence–chemistry interactions are important. That is the case in these syngas flames, because of the slow CO chemistry. The Damköhler number is estimated to be unity at \( x/d = 20 \) for flame B (Barlow et al., 2000), and decreases with downstream distance. Therefore, these syngas flames tend to have broad reaction zones (Chen et al., 1991), and finite-rate chemistry plays an important role.

Results obtained using two different mixing models (MC and EMST) were compared first for the same value of \( C_{\phi} (C_{\phi} = 1.5) \), and only minor differences were found (not shown). Then results obtained using EMST were compared for different values of \( C_{\phi} \): 1.0, 1.5, 2.0, and 8.0. Increasing \( C_{\phi} \) increases the mixing rate; in the

![Figure 6](image_url)  
*Figure 6* Comparisons of results obtained using three different chemical mechanisms at \( x/d = 40 \) for flame A. (a) Mean CO₂; (b) mean H₂. (Figure is provided in color online.)
Figure 7 Comparisons between full GRI-2.11 NOx chemistry and GRI-2.11 with extended Zeldovich thermal NO only for flame A. (a) Mean NO mass fraction, $x/d = 20$; (b) mean NO mass fraction, $x/d = 40$. (Figure is provided in color online.)

limit $C_{\phi} \rightarrow \infty$, local fluctuations go to zero, and the model essentially reduces to a well-stirred reactor model. While $C_{\phi} = 8.0$ is high compared to values that normally are used in PDF-based modeling studies, it serves to illustrate the trends and the

Figure 8 Comparisons of results obtained using different values of $C_{\phi}$ for flame B. (a) Mean temperature, $x/d = 20$; (b) rms temperature, $x/d = 20$; (c) mean NO mass fraction, $x/d = 20$; (d) rms NO mass fraction, $x/d = 20$; (e) mean temperature, $x/d = 40$; (f) rms temperature, $x/d = 40$; (g) mean NO, $x/d = 40$; (h) rms NO, $x/d = 40$. (Figure is provided in color online.)
importance of turbulent fluctuations. Examples are shown in Figure 8. The changes in computed mean and rms profiles for values of $C_\phi$ between 1.0 and 2.0 are relatively small, but they are dramatic with $C_\phi = 8.0$. For $C_\phi = 8.0$, local composition and temperature fluctuations are damped significantly, and the computed peak mean temperature increases by over 200 K. This underscores the importance of accounting properly for turbulence-chemistry interactions in these flames.

**Flame Structure in Mixture Fraction Space**

Conditional mean temperature and OH mass fractions obtained using two mixing models (MC and EMST with $C_\phi = 1.5$, all other values correspond to the baseline model) are compared with measurements in Figure 9 for flame B. The definition of the mixture fraction and the mixture fraction bin width are consistent with those reported in the experiments (Barlow et al., 2000). The two mixing models give similar profiles, although the MC results show more fluctuations. The conditional mean temperature profiles show good agreement with experiment at $x/d = 20$ and on the fuel-lean side at $x/c_l = 50$. However, the conditional mean OH mass fractions are overpredicted at both locations with both mixing models. With consideration of radiation, the peak mean temperatures would drop by as much as 100 K. The prediction of minor species on the fuel-rich side is expected to improve with better chemical mechanisms. Hewson and Kerstein (2001) also showed conditional mean temperature

![Figure 9](image)

**Figure 9** Comparisons of conditional mean profiles obtained using different mixing models for flame B. (a) Conditional mean temperature, $x/d = 20$; (b) conditional mean OH mass fraction, $x/d = 20$; (c) conditional mean temperature, $x/d = 50$; (d) conditional mean OH mass fraction, $x/d = 50$. (Figure is provided in color online.)
and species mass fraction profiles. In their results, the conditional mean temperatures at both location were overpredicted, while the conditional mean OH predictions showed good agreement with the experiment.

Scatter plots of temperature versus mixture fraction for flame B obtained using the EMST mixing model with $C_\phi = 1.5$ can be found in the Supplementary Material online. Local extinction was reported by Hewson and Kerstein (2001) at $x/d = 6$, where no experimental data are available. Here, no local extinction is found at $x/d = 6$, $x/d = 20$ and $x/d = 50$, which is consistent with the experimental observations.

CONCLUSION

A consistent hybrid Lagrangian particle/Eulerian mesh composition PDF method has been used to simulate two turbulent syngas flames. Key elements of the baseline model include the use of a standard $k-\varepsilon$ turbulence model, gradient transport for scalars, and the EMST mixing model. The baseline model reproduces the measured mean and rms temperature, major species, and minor species profiles reasonably well, and captures the scaling that is observed in the experiments. Further improvements can be realized with adjustments in the current turbulence model or using an alternative model, consideration of radiation heat transfer, and improved chemical mechanisms. Although radiation effects are relatively small in these flames, consideration of radiation is important for accurate NO predictions. In the oxyfuel context, NO is more of an issue for potential corrosion in the CO$_2$ pipelines (via acid formation), rather than as an air pollutant. Chemical mechanisms that have been developed specifically for syngas (high concentrations of fuel CO and H$_2$) perform better than a benchmark mechanism (GRI-Mech 2.11) that was not designed for this purpose. It is important to account explicitly for turbulence-chemistry interactions, although the details of the mixing model do not make a large difference, within reasonable limits. Remaining discrepancies between model and experiment may, in part, be attributed to the neglect of differential diffusion.

In future work, we will move toward environments that are more representative of the target oxy-coal application. Accurate treatment of radiation heat transfer will be more important, and the spectral radiation model will be extended to include CO.

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