PREDICTION OF NO\textsubscript{x} FORMATION IN A PRACTICAL-SCALE, NONPREMIXED TURBULENT FLAME USING EDDY DISSIPATION AND RADIATIVE FLAMELET COMBUSTION MODELS

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A CFD-based combustion model coupled with a NO\textsubscript{x} postprocessing model has been applied to simulate a practical-scale, turbulent nonpremixed natural gas flame studied experimentally in a 3 MW furnace at the International Flame Research Foundation (Michelfelder and Lowes, 1974). The combustion is modeled using a radiative flamelet combustion (RFC) model (Ma et al., 2002), which is an extension of the mixedness-reactedness flamelet combustion model of Bradley et al., (1990) and the eddy-dissipation combustion (EDC) model of Magnussen and Hjertager (1976). To account for the radiation heat transfer, a comprehensive radiation model, namely, the discrete transfer method of Lockwood and Shah (1980), has been introduced into the combustion code. The turbulence is represented by the $k$–$\varepsilon$ model with the standard values of the constants and variants of $C_\varepsilon$. The predicted velocity, temperature, and major species concentration fields obtained from both combustion models have been used to calculate nitric oxide (NO) formation in the furnace. The computational results are compared with experimental data for gas temperature, oxygen, and NO concentrations. The EDC model is unable to capture the effects of finite-rate reactions and hence fails to predict the flame structure in the near burner region causing the overpredictions of NO concentrations in this region. The RFC model reproduces the overall structure of the flame reasonably well and shows better agreement of the predicted NO concentrations with measurements.

Keywords: NO prediction, Combustion modeling, Radiation, Industrial-scale flame

INTRODUCTION

In recent years, computational fluid dynamics (CFD) models have commonly been used for predictions of emission levels of NO\textsubscript{x} produced by industrial combustion equipment and in investigating methods to keep these levels down to a value that complies with legislation while maintaining good combustion efficiency. The accuracy of the calculated NO\textsubscript{x} levels depends, in addition to the near burner aerodynamics/mixing, on how well the temperature and oxygen concentration distributions are predicted. Many of the burners employed in industrial combus-

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tion equipment are based on nonpremixed combustion and designed in such a way that rapid initial mixing of fuel and air streams occurs. No flame can exist at aerodynamic strain rates above a critical value due to quenching effects. However, when the strain rate reduces to a value where ignition can take place, appreciable premixing of the fuel and air streams occurs upstream of the location where the flame is stabilized. In predicting burner performance, and to assist in the design of efficient, low-NO\textsubscript{x} burners, there is therefore a requirement for accurate mathematical models of turbulent nonpremixed combustion.

A variety of combustion modeling approaches is available in the literature [see review by Libby and Williams (1994), Veynante and Vervisch (2002)]. For the calculation of turbulent nonpremixed flames in industrial combustion equipment, equilibrium combustion models are generally employed. These models are unable to capture the important effects of finite-rate reactions such as ignition, extinction, flame structure, and stability. In order to enhance the quality of predictions, a further approach has been the use of a stretched laminar flamelet concept, based on either diffusion (Bray and Peters, 1990) or premixed (Bradley \textit{et al.}, 1990, 1998) flamelets. It is important to note that in the flamelet modeling approach, the assumption of adiabatic combustion is generally invoked, which restricts the applicability of the model to situations with negligible radiation heat losses. It is well known that thermal radiation plays an important role in industrial flames. In our previous study (Ma, 2002), the adiabatic mixedness-reactedness flamelet combustion model of Bradley and coworkers (Bradley \textit{et al.}, 1990, 1998) has been extended to incorporate the radiative heat loss effect, based on the optically thin medium assumption, using the concept of enthalpy defect (Bray and Peters, 1994). Computed results of lifted natural gas jet flames, studied experimentally by Lockwood and Moneib (1982) and Eickhoff \textit{et al.} (1984), obtained with the original adiabatic flamelet model (Bradley \textit{et al.}, 1990), show significant overprediction of the mean gas temperature in the downstream region of the flame. The inclusion of radiation heat loss brings predictions of the flame structure more in line with the observed structure of lifted, nonpremixed flames. This approach has also been implemented by a few authors in the field to account for radiative heat loss from nonpremixed flames [see, for example, (Marracino and Lentini, 1997; Brooks and Moss, 1999; Hossain \textit{et al.}, 2001); Mateus \textit{et al.}, 2004].

The heat loss due to radiation appears to be often overlooked in many previous turbulent combustion modeling studies using laminar flamelet approaches, as noted by Marracino and Lentini (1997) and Hossain \textit{et al.} (2001). When radiation was considered, simplified assumptions were made in order to account for the heat loss (Chen and Chang, 1996), which may be appropriate when considering small-scale flames. However, for large-scale flames in practical combustors, the
radiation heat loss becomes more significant and requires a comprehensive radiation modeling approach. In a recent study, Wen and Huang (2000) considered two large-scale confined propane jet fires under ventilation-controlled conditions. They applied three different combustion models, including a flamelet model, coupled with the discrete transfer method (DTM) of Lockwood and Shah (1980) for the radiative heat loss. The predictions show that the flamelet model produces the best agreement with measurements, and demonstrates the importance of accounting for the radiative heat exchange in the calculations of such large-scale flames. However, applications of flamelet models coupled with radiative heat transfer to practical flames are rare in the available literature.

In the present study, a comprehensive radiation model, namely, the DTM (Lockwood and Shah, 1980), has been introduced into our radiative flamelet combustion (RFC) model (Ma et al., 2002) because the optically thin flame assumption is not adequate for practical-scale flames. The RFC model and the commonly used eddy-dissipation combustion (EDC) model of Magnussen and Hjertager (1976) have been employed to simulate a turbulent nonpremixed natural gas jet flame in a large-scale experimental furnace with 3 MW thermal input at the International Flame Research Foundation (IFRF), IJmuiden (Michelfelder and Lowes, 1974). The predicted velocity, temperature, and major species concentration fields obtained from both combustion models together with the standard $k-\varepsilon$ turbulence model have been used to calculate nitric oxide (NO) formation in the furnace. NO predictions are obtained using the Zeldovich mechanism for thermal-NO (Zeldovich et al., 1947) and the kinetic rate expression of De Soete (1975) for the formation of prompt-NO. The effect of turbulence-chemistry interactions on NO formation rates is represented by a single variable beta probability density function (PDF). Predictions are compared with the measured temperature and oxygen concentration, and also with the in-flame and flue-gas NO concentrations.

**MATHEMATICAL MODELS**

**Aerodynamic/Combustion Model**

The present mathematical model is based on the numerical solution of the Favre-averaged conservation equations for mass, momentum, thermal energy, and chemical species. The turbulent momentum flux is represented by the standard $k-\varepsilon$ turbulence model and the scalar fluxes by a gradient-diffusion type model with a constant turbulent Prandtl/Schmidt number (Lauder and Spalding, 1974). The source terms in the major chemical species and thermal energy conservation equations representing the mean turbulent combustion reaction and heat release rates,
respectively, are obtained using the RFC and EDC models. It should be noted that the EDC model retains the original values of the constants.

The natural gas fuel used in the IFRF furnace (Michelfelder and Lowes, 1974) was represented by pure methane in the calculations. For the RFC model, the laminar heat release rate, \( q_l(c, \theta, q_r) \), and the reaction rate of species \( i, w_{i,l}(c, \theta, q_r) \), are expressed as a function of the mixture fraction \( c \), a reaction progress variable \( \theta \), and the radiative heat loss \( q_r \) (Bradley et al., 1990; Ma et al., 2002). A nonadiabatic flamelet data library comprising the reaction and heat release rates was generated (Ma et al., 2002) from detailed calculations of one-dimensional laminar premixed, methane-air flames with the GRI (2.11) chemical reaction mechanism (Bowman et al., involving 49 species and 279 reaction steps, using the computer code CHEMKIN (Kee et al., 1993). The turbulent mean values of \( q_l \) and \( w_{i,l} \) can be obtained using the joint PDF of \( c, \theta, \) and \( q_r \). In the present calculations, statistical independence between these parameters was assumed and the effect of fluctuations of \( q_r \) was neglected. Consequently, the joint PDF was represented by the product of independent beta function PDFs of \( c \) and \( \theta \), and a delta function for \( q_r \).

The source term in the thermal energy equation representing radiative heat transfer was calculated using the DTM of Lockwood and Shah (1980). This method uses a recurrence relation to calculate the radiative heat transfer along a number of specified paths. Along each of these paths, the radiation intensity entering and leaving each computational cell is calculated for each ray, which then allows the source term to be calculated. An estimated value of the initial intensity is used to start the iterative calculations at the walls in order to trace the representative rays from one surface to another through the intervening cells. The properties within each cell are assumed to be uniform. The temperature and emissivity of the IFRF furnace walls were specified from measured data reported by Michelfelder and Lowes (1974). The absorption coefficients of radiating species were obtained from the gray-gas model of Truelove (1976). The presence of soot particles in the radiation calculations was neglected because a clear blue natural gas flame was observed in the furnace (Michelfelder and Lowes, 1974).

An existing CFD code, described in detail by Ma et al. (2002), has been used to solve the governing conservation equations. This code was originally developed by Bradley and coworkers (Bradley et al., 1990; Gu, 1993) to calculate the velocity and temperature fields using the adiabatic mixedness-reactedness flamelet combustion model, and later developed further by Mahmud and coworkers to include the EDC model and species conservation equations (Ma et al., 1999), and radiative heat transfer based on the optically thin flame assumption (Ma et al., 2002).
NO Models

The nitric oxide (NO) concentration is calculated using a CFD-based postprocessing NO model developed by Ma et al. (2000). The accuracy of NO concentration predictions does not only depend on the precision of the aerodynamic/combustion predictions, but also on the chemical reaction mechanisms used to represent the formation and destruction of NO and the turbulence-chemistry interactions. For the combustion of natural gas, in the absence of any nitrogen-containing compounds, the two main sources that account for the formation of NO in combustion equipment are: thermal-NO (oxidation of atmospheric nitrogen) and prompt-NO (reactions of hydrocarbon free radicals with atmospheric nitrogen).

The formation of thermal-NO is modeled using the Zeldovich mechanism including the reverse reactions. A partial equilibrium assumption is employed for the calculation of the oxygen-atom concentration. The rate constants were obtained from Baulch et al. (1973). It is worth noting that this assumption may not be valid at temperatures below 1500 K (Drake, 1986). However, in the IFRF furnace, temperature levels within the combustion zone exceed this value and the assumption of partial equilibrium is therefore adequate. The most common method for the calculation of the oxygen-atom concentration has been the use of the partial equilibrium assumption for enclosed practical-scale flames where this approach was found to be adequate (Boardman et al., 1993; Coelho and Cravalho, 1995; Williams et al., 1997; Ma et al., 2000; Falcitelli et al., 2002) and is therefore used in the present study. It should be noted that the oxygen-atom concentration can also be obtained from the flamelet data (Benim and Syed, 1998); however, as shown in a recent investigation carried out by Hossain and Malalasekera (2001), the flamelet approach has difficulties in predicting the experimental NO levels.

The reaction mechanism for the production of NO via the prompt-NO route is complex and hence a global rate expression proposed by De Soete (1975) for the combustion of hydrocarbon fuels is used in this study. The NO concentration distribution is obtained by solving the Favre-averaged species conservation equation using the predicted velocity, temperature, and major species concentration data obtained from the aerodynamic/combustion model. The mean turbulent NO reaction rates are obtained, taking into account the effect of temperature fluctuations only, using a beta-function PDF. This approach has been widely used in previous computational studies, for example, by Ma et al. (2000), Peters and Weber (1995), and Boardman et al. (1993) for practical flames and was found to be adequate.
SIMULATION OF IFRF FURNACE

In this study, the experimental data for Flame-29 of M-2 trials at IFRF (Michelfelder and Lowes, 1974) was used to validate the model predictions. Flame-29, which has a 3 MW thermal input, is a nonswirling turbulent natural gas (81.3% methane) flame produced by a double-concentric cylindrical burner. The combustion chamber of the IFRF furnace is square in cross section and the length and width of the chamber are 6.25 m and 2.26 m, respectively, as shown schematically in Figure 1. In the computation, the furnace was converted to a cylindrical one with an equivalent cross-sectional area for corresponding cylindrical coordinates. The experimental conditions, which were specified as the input conditions for the calculations, are given in Table 1.

The computations were carried out on a staggered, two-dimensional, nonuniform mesh containing 49 (radial) \( \times \) 55 (axial) grid lines, with a higher grid density in the region near the burner exit and the furnace axis. Test calculations with different mesh sizes, 22 \( \times \) 22 and 106 \( \times \) 48, showed that the selected mesh was sufficiently fine to provide acceptable grid-independent solutions. The input data for the calculations was obtained from measurements where available.

![FIGURE 1 IFRF experimental furnace.](image)

<table>
<thead>
<tr>
<th>TABLE 1 IFRF furnace operating conditions for Flame-29</th>
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<tr>
<td>Excess oxygen flow rate: 170 kg/hr</td>
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<tr>
<td>Air/Oxygen inlet velocity: 36 m/s</td>
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<tr>
<td>Air temperature: 26°C</td>
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<tr>
<td>Fuel flow rate: 280 kg/hr</td>
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<tr>
<td>Fuel inlet velocity: 112 m/s</td>
</tr>
<tr>
<td>Fuel temperature: 10°C</td>
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<tr>
<td>Air flow rate: 3126 kg/hr</td>
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RESULTS AND DISCUSSION

Flame Predictions

Figure 2 shows comparisons between the predicted radial profiles of mean gas temperature and oxygen concentration and the experimental data (Michelfelder and
Lowes, 1974) at different axial distances from the burner exit. The predictions were obtained using both the EDC and RFC models together with the standard $k-\varepsilon$ model for turbulence. At stations $x = 0.37, 1.48,$ and $2.035$ m (where $x$ is the axial distance from the burner exit), the EDC model significantly overpredicts the gas temperatures and underpredicts the oxygen concentration levels around the furnace axis. This suggests early ignition and hence fails to capture the experimentally observed structure of the lifted flame in the near burner region. As for the RFC model, the predicted gas temperature and oxygen concentration in the nonreacting lift-off region of the jet (at $x = 0.37$ m) are in good agreement with the experimental data, indicating that the onset of combustion is correctly simulated. The predicted off-axis peak temperatures within the combusting zone of the jet, at $x = 1.48, 2.035,$ and $2.75$ m, are in reasonably good agreement with measurements. However, discrepancies exist at and about the furnace axis where the predicted temperatures are much lower and oxygen concentrations are much higher compared with measurements. Calculations were also performed using the optically thin flame assumption for radiation heat transfer as well as the adiabatic flamelet model without accounting for radiative heat losses. These two approaches resulted in significant overpredictions of the gas temperature.

Figures 3 and 4 show contours of the predicted turbulent mean heat release rates and corresponding gas temperatures for 4 m along the length of the furnace using the EDC and RFC models, respectively. The contour of heat release at a rate of $5 \times 10^6$ W/m$^3$ is taken as the flame boundary, as suggested by Bradley et al. (1990) and Ma et al. (2002). These figures reveal overall structures of the flame as predicted by the two combustion models. Figure 3, which represents the temperature distribution and the heat release rates predicted by the EDC model,
shows early ignition of the flame. In contrast, the RFC model predictions (see Fig. 4), in accordance with the experimental observation, reveal a lifted flame. The extended low-temperature central core around the furnace axis suggests that ignition is suppressed there.

As depicted in Figure 2, the RFC model underpredicts the gas temperatures and overpredicts the levels of oxygen concentration in the central core of the jet at \(1.48 \leq x \leq 2.75\) m, indicating that ignition is suppressed in this region. This may be attributed to the excessive strain rates resulting from the overestimation of velocity at the centerline of the jet. The mean axial velocity along the centerline is sensitive to the spreading rate of the jet, and previous studies, for example, Pope (1978) and Van Slooten and Pope (1999), have shown that large errors can occur when predicting the spreading rate of an axisymmetric jet issuing into a coflowing stream or stagnant environment using the standard values of the \(k-\varepsilon\) turbulence model constants. There have been several modifications to the \(k-\varepsilon\) model constants in order to rectify this inaccuracy (Pope, 1978; Gran et al., 1997; Van Slooten and Pope, 1999; Prasad et al., 1999; Dally et al., 1998; Roerkaerts et al., 2000). It is well known that the standard model constants overpredict the jet spreading rate and hence the velocity decay rate. In order to compensate for this deficiency, most authors have suggested a value of 1.60 for the constant \(C_{\varepsilon_1}\). However, less commonly seen in the literature is a case where the spreading rate of the jet needs to be increased in order to increase the rate of decay of the centerline velocity. Van Slooten and Pope (1999) investigated the effect of values of \(C_{\varepsilon_1}\) when calculating confined swirling and nonswirling coaxial jets. For a very mildly swirling jet (swirl number = 0.09), the value of \(C_{\varepsilon_1}\) was reduced from the standard value of 1.44 to 1.32.
FIGURE 5 Comparison between predicted and measured temperatures and O$_2$ concentration distributions using the RFC model (—, standard k-ε model; - - - , $C_{e1} = 1.60$; ---, $C_{e1} = 1.32$; △, measurement).
FIGURE 6 Comparison between predicted and measured temperatures and O₂ concentration distributions using the RFC model (——, Pr/Sc = 0.5; ----, Pr/Sc = 0.2; Δ, measurement).
In the present study, the sensitivity of the predictions to variations of the turbulence model constant $C_{\varepsilon 1}$ was examined. Figure 5 shows the predicted radial temperature and oxygen concentration profiles at selected stations along the length of the furnace obtained using the RFC model and three values of $C_{\varepsilon 1}$. When comparing the predicted temperature profiles obtained using $C_{\varepsilon 1} = 1.60$ to those that for the standard value, $C_{\varepsilon 1} = 1.44$, it is evident that increasing the value of $C_{\varepsilon 1}$ has resulted in the reduction of the jet spreading rate and hence the centerline velocity decay rate. However, decreasing the value of $C_{\varepsilon 1}$ from 1.44 to 1.32 produces an opposite effect. As a result, the strain rates are relaxed, causing the flame front around the centerline to move toward the burner. This has resulted in a better agreement between the predictions and experimental data at $x = 2.75$ m. However, discrepancies still exist at stations $x = 1.48$ and 2.035 m. It appears that the value of $C_{\varepsilon 1}$ needs to be reduced further in order to improve the quality of predictions.

As mentioned before, a gradient-diffusion type model with a constant Prandtl (Pr) and Schmidt (Sc) number for the turbulent scalar fluxes has been used in this study. In order to enhance the rate of mixing between the central fuel jet and the coflowing air jet, Pr and Sc were reduced to 0.2. As shown in Figure 6, the quality of the predicted temperature and oxygen concentration has improved in the vicinity of the furnace axis, but at the expense of the quality of predictions in the outer region of the jet. From the above sensitivity analyses, it appears that instead of adjusting the turbulence models’ constants on an ad hoc basis, a better approach would be to employ second-order closure models for the turbulent momentum and scalar fluxes.

**NO Predictions**

Figure 7 shows the predicted radial profiles of NO concentration obtained using the aerodynamic/combustion results produced from both combustion models together with the available experimental data. As can be seen, the predicted levels of NO concentration around the furnace axis in the region $x \leq 2.75$ m obtained using the EDC model are significantly higher than the measured values. The predictions also reveal that NO formation starts in the proximity of the burner (at $x = 0.37$ m), which corresponds to high temperatures resulting from the early ignition predicted by this combustion model. It is apparent that the high on-axis peak temperatures produced by the EDC model dominate the formation of NO in this region. As for the RFC model, the predicted NO concentrations at the furnace axis up to a distance $x \leq 2.75$ m are in better agreement with experimental measurements. However, this may be fortuitous due to the underestimation of temperature in this region. The off-axis peaks in the predicted NO concentration profiles at $x = 1.48, 2.035,$ and 2.75 m correspond to the peaks in the predicted tempera-
ture profiles at the same locations, as shown in Figure 2. The level of NO concentration near the furnace exit, at $x = 5.75$ m, obtained using the RFC model is in good agreement with the measurement compared with the EDC model.

The NO concentration distribution and its rate of formation predicted using the aerodynamic/combustion results obtained from the EDC and RFC models are presented in Figures 8 and 9, respectively. For the case of the EDC model, the NO formation mainly occurs in the region that extends up to an axial distance of about $x = 1.8$ m. Although the combustion zone, as shown in Figure 3b, extends up to $x = 3.4$ m, the NO formation rates are very small in the downstream region beyond $x > 1.8$ m. This is because the predicted oxygen concentration levels around the furnace axis are virtually zero, as revealed in Figure 2. The rates of NO formation predicted in conjunction with the RFC model show a much longer reaction zone, which corresponds to the length of the combustion zone where oxygen concentrations are high relative to the EDC model predictions.
CONCLUSIONS

A nonadiabatic radiative flamelet combustion (RFC) model coupled with the discrete transfer method for radiation heat transfer has been applied to calculate a turbulent nonpremixed natural gas flame in a large-scale experimental furnace at IFRF. Calculations were also performed using the widely employed eddy-dissipa-
tion combustion (EDC) model. The predicted velocity, temperature, and major species concentration fields obtained from both combustion models together with the standard $k-\varepsilon$ turbulence model have been used to calculate NO formation in the furnace.

The present study demonstrates that a stretched laminar flamelet model with radiative heat transfer leads to encouraging results that closely reproduce the overall structure of a practical-scale flame and its emission characteristics. However, the RFC model predicts delayed ignition in the central core region of the jet resulting from the overestimation of the strain rates. This predictive difficulty is primarily linked to the deficiencies of the eddy-viscosity based models for the turbulent transport of momentum, heat, and mass, as revealed by the sensitivity study effected in order to examine the influence of model constants on the predictions in this region. Therefore, further investigation is required by employing second-order closure models for the turbulent transport processes. The more widely used EDC model with the original constants fails to capture the measured trends in the near burner region. The gas temperature is overpredicted in this region, resulting in significantly high levels of NO concentration. The present investigations emphasize the need for an accurate prediction of the flame structure in order to achieve reliable predictions of NO emissions from industrial combustors.

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