



TRANSPORT AND CHEMICAL KINETICS OF H₂/N₂ JET FLAME: A FLAMELET MODELLING APPROACH WITH NO_x PREDICTION

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Abstract

In this work simulation of a turbulent H₂/N₂ jet diffusion flame with flamelet modelling has been presented. The Favre-averaged mixture fraction has been employed to model the combustion. Favre-averaged scalar quantities have been calculated from flamelet libraries by making use of a presumed Probability Density Function (PDF) method. The predicted flame temperature profiles and chemical species concentrations are compared with TNF experimental data obtained from Sandia/California. Predictions considering the unity Lewis number flamelet are found to be in good agreement with temperature and chemical species measurements. Predicted NO results also compared and shown with other species. This study shows that the combustion simulation using unity Lewis number flamelets are effective for predicting the flow, temperature and chemical kinetics of H₂/N₂ diffusion flame. To account for fluctuations of mixture fraction, its distribution is presumed to have the shape of a beta-function.

Keywords: Non-premixed flame, Turbulent Combustion, Flamelet modelling

1. Introduction

Combustion is the oldest and fundamental technology of mankind; it has been used for more than one million years and the mainstay of worldwide industrial development for the past 200 years. At present, about 90% of our worldwide energy supports in different sectors like transport, electrical power generation, heating are provided by combustion; Also the combustion systems used in power generation and transportation industries, to generate steam and heat for other vital manufacturing processes and to change the mechanical and chemical properties of materials and products are growing rapidly. This induces pollution and environmental problems to become critical factors in our societies. Environmental issues such as ozone transport and global climate change are emerging as defining factors in the design and operation of combustion equipment. The good predictions of temperature and major species are no longer sufficient, the combustion model should also be able to predict minor species, radicals and emissions of pollutants. The growing concerns about environmental pollution and the strict regulations regarding pollutant emissions make it necessary to find a suitable predictive tool for emissions calculation. The prediction of NO poses the most stringent test of the combustion models, because the NO production rate is very sensitive to the accurate prediction of temperature and concentration of O. It is also important to study the influence of radiation heat transfer on the temperature and minor species but in this paper, the flamelet based NO prediction studies are undertaken and the consideration of radiation heat transfer have been left for future investigation.

Several combustion models that account for the interaction of turbulence and chemistry have been developed and applied to a number of flames ranging from simple jet flames to complex combustion chambers. For reliable predictions, advanced combustion models need to be employed in the calculation of industry-relevant flames where finite-rate chemistry and non-equilibrium effects are often significant. One such approach to modelling turbulent non-premixed flames has been the use of stretched laminar flamelet models based on diffusion flamelets can be found in Peters (1984,1986). Among the currently available combustion models the other successful models are the conditional moment closure (CMC) developed by Bilger (1993) and probability density function (PDF) transport

model by Pope (1990, 1995). The present study is concerned with the flamelet model and the application of the flamelet model for NOx prediction. The use of the flamelet model for the prediction of NO has begun only in the last few years by Sanders and Gökalp (1995), Schlatter *et al* (1996) and Sanders *et al* (1997) and. The work by Schlatter *et al* (1996) and Sanders and Gökalp (1995) and Sanders *et al.* (1996) showed that the scaling behaviour of NO emission was not reproduced when the strain rate was used as the non-equilibrium parameter. Chen and Chang (1996) have investigated the effects of differential diffusion and radiation on NO formation. According to their findings, the inclusion of radiation heat transfer reduced the predicted level of NO. Predictions are considered with unity Lewis number flamelet and the flamelet model with differential diffusions. Predicted NO results also compared with the experimental data and shown with the other species. The H₂/N₂ jet diffusion flame experimental data can be accessed through from Sandia/California TNF WebPages.

2. Model Formulation

2.1 Flamelet Modelling

According to the laminar flamelet concept, as presented in previous reviews by Peters (1984) and by Bray and Peters (1994), a turbulent non-premixed flame is considered to be comprised of an array of laminar ones. A conserved scalar variable, the mixture fraction, is then introduced to describe the flame structure, with the thermochemical state variables and mixture fraction relationships obtained either from laminar flame measurements by Moss *et al* (1988) or by performing calculations of one-dimensional, adiabatic counter-flow laminar diffusion flames with a detailed chemical kinetic scheme, as a function of the strain rate. Mean values of the thermochemical state variables, such as gas density, species concentrations and temperature, can then be evaluated using a joint probability density function (PDF) of the mixture fraction and strain rate, which is frequently decomposed into a product of presumed individual PDFs. Peters (1984) presented an extensive review of flamelet approach for modelling turbulent combustion. By transformation of the physical coordinate into one with the mixture fraction and under the thin flame assumption, it was shown by Peters (1984) that in the steady state, temperature T , and species mass fraction Y_k are determined by the balance between diffusion and chemical reaction as:

$$\rho \frac{\chi}{2} \frac{d^2 T}{df^2} = \frac{1}{C_p} \sum_{k=1}^N \dot{\omega}_k h_k \quad (1)$$

and

$$\rho \frac{1}{Le_k} \frac{\chi}{2} \frac{d^2 Y_k}{df^2} = -\dot{\omega}_k, k = 1, 2, \dots, N \quad (2)$$

where f is mixture fraction, $\chi = 2\alpha \nabla f \cdot \nabla f$ is the scalar dissipation rate, $\dot{\omega}_k$ is the chemical source term, C_p is the mean mixture specific heat, $\alpha = \lambda / \rho C_p$ is the thermal diffusivity, λ is the thermal conductivity, $Le_k = \alpha / D_k$ is the Lewis number of the k -th species, and D_k is the diffusion coefficient of species k . The boundary conditions for flamelet equations 1 and 2 are:

$$T(f = 0, \chi) = T_{\text{oxidizer}}, T(f = 1, \chi) = T_{\text{fuel}} \quad (3)$$

$$Y_k(f = 0, \chi) = Y_{k,\text{oxidizer}}, T(f = 1, \chi) = Y_{k,\text{fuel}} \quad (4)$$

The fuel properties are specified by the product gases in the downstream level. For turbulent flames, the mean scalar variables are computed from the laminar flamelet relation of the mixture fraction and the scalar dissipation rate by integration over a joint probability density function as:

$$\tilde{\phi} = \int_0^{\infty} \int_0^1 \phi(f; \chi) P(f) P(\chi) df d\chi \quad (5)$$

The assumption of statistical independence leads to $P(f, \chi) = P(f) P(\chi)$ as suggested by Peters (1984). The Favre average PDFs of mixture fraction, $P(f)$ is assumed to be a beta distribution and that the

scalar dissipation rate, $P(\chi)$ is assumed to be a log-normal distribution after Peters (1984). In CFD code transport equations are solved for the mean mixture fraction, \tilde{f} and mixture fraction variance, \tilde{f}^{n2} . The mean scalar dissipation rate in the turbulent flow field can be modelled according to:

$$\tilde{\chi} = C_{\chi} \frac{\tilde{\varepsilon}}{\tilde{k}} \tilde{f}^{n2} \quad (6)$$

where C_{χ} is a constant with a value of 2.0 Bray and Peters (1994). The width of the log-normal distribution is given in terms of variance and is assigned as $\sigma_{\chi}^2 = 2.0$ after Peters (1984). The solution of above flamelet equations for given concentration and temperature boundary conditions, and various χ provides a flamelet library $Y_k(f, \chi)$. A variety of techniques are available to build these libraries and the flamelet libraries are generated using flamelet code RUN1-DL by Rogg (1995).

2.2 NOx Modelling

Strong dependence of the NO_x formation rate on gas temperature and the amount of nitrogen in the fuel have been emphasised in many studies. Both characterise the most important routes for the formation of NO_x. It become apparent that nitric oxide (NO) and nitrogen dioxide (NO₂), collectively called NO_x. Normally NO is formed in much larger amounts than NO₂, and the latter is thought to be formed by further reaction of NO. Hence, NO formation determines the total amount of NO_x emitted. Combustion generated pollutants like NO are formed near the reaction zone. Since the formation rates of pollutant species are relatively slow compared to the combustion, the concentration of the pollutant species depend strongly on flow residence time. Pollutant concentrations can not be obtained directly from the steady-state flamelet library or from the equilibrium chemistry. The present numerical scheme solves for the species conservation equation of NO directly during the simulation. The slow, kinetically limited production of NO is obtained by solving the transport equation for NO

$$\frac{\partial}{\partial x_j} \bar{\rho} \tilde{u}_j \tilde{Y}_{NO} = \frac{\partial}{\partial x_j} \left(\frac{\mu_{\text{eff}}}{\sigma_{NO}} \frac{\partial Y_{NO}}{\partial x_j} \right) + \bar{\omega}_{NO} \quad (7)$$

where σ_{NO} is the turbulent Schmidt number, which is taken as 0.7. For a given (f, χ) state, the source term for NO is evaluated by the corresponding chemistry states of major and intermediate species from the flamelet library. Since local NO concentration will be needed in evaluating the reverse rate, the NO source term split into two parts

$$\dot{\omega}_{NO} = \dot{S}_{NO} - [NO] / \tau_{d-NO} \quad (8)$$

Where the first term, \dot{S}_{NO} represents the production rate of thermal NO and it is positive. The second coefficient, τ_{d-NO} , is the time scale of NO destruction. The coefficients \dot{S}_{NO} and τ_{d-NO} in equation (8) are determined by extended Zeldovich mechanism and the reburning reactions with the steady-state assumption for the atomic nitrogen. The mean rate, $\bar{\omega}_{NO}$ is obtained from the following formula

$$\bar{\omega}_{NO} = \bar{\rho} \tilde{\omega}_{NO} = \bar{\rho} \int_0^1 \int_0^{\infty} \dot{\omega}_{NO}(f, \chi) P(f) P(\chi) df d\chi \quad (9)$$

Modelling of NO formation by Bartok *et al.* (1971) shows the effect of several variables:

- higher residence times at higher temperatures yield higher NO.
- maximum formation of NO occurs at 5 % excess air, and NO levels drop off at 10 % excess air.
- increase of preheat temperature has dramatic effect by increasing NO.

Four different routs are identified in the formation of NO_x by Bowman (1996). These are the thermal NO route, the prompt NO route, the N₂O (nitrous oxide) route, and the fuel-bound nitrogen route. Thermal NO formed by high-temperature oxidation of atmospheric nitrogen. Thermal NO or

Zeldovich-NO is formed by well-known Zeldovich mechanism and is enhanced by presence of superequilibrium O and OH and is very sensitive to temperature. This mechanism has been considered here in this study. In the prompt NO mechanism, hydrocarbon fragments attack the bimolecular nitrogen, producing atomic nitrogen, cyanides and amines, which are subsequently oxidized to NO. The N_2O route is analogous to the thermal route in that the O atom attacks the molecular nitrogen. However, with the presence of a third body, N_2O is formed and this subsequently reacts with O atoms to reduce NO. The N_2O route is unimportant at ambient pressure. The relative contribution from different pathways depends on fuel type, temperature, pressure and residence time as described by Bartok *et al.* (1971). The high NO levels that occur in practical systems can only be reduced by reducing the thermal NO formation. The thermal NO is modelled by the Zeldovich mechanism. The reaction rate parameters for the Zeldovich mechanism, adapted here are well known and are explained extensively by Warnatz *et al.* (1996).

3. Numerical Solution

The discretization of the governing transport equations are performed by using SIMPLE algorithm together with TDMA given by Patankar (1980). The k - ϵ turbulence model is used for turbulence closure in the present study. The standard k - ϵ model has known shortcomings for predicting round jets. In particular, the k - ϵ model overpredicts the decay rate and the spreading rate of a round jet. Several modifications to the k - ϵ model are available for correcting this problem McGuirk and Rodi (1979). In the present study, a simple modification to the value of $C_{\epsilon 1}$ of the ϵ -transport equation is made. The value of constant $C_{\epsilon 1}$ is modified from 1.44 to 1.60 following the work of McGuirk and Rodi (1979) and Dally *et al.* (1998). Significant improvement were found in the results obtained from their computations using the new constant and validation of the modification of $C_{\epsilon 1}$ were made by applying to the jet flame problems.

4. Results and Discussion

4.1 Burner and jet configurations

The flame considered here consists of 75% H_2 + 25% N_2 by volume and has a jet velocity of 42.3 m/s with $Re=9300$ and the simulated results have been shown in figures 1 and 2. The experimental data for the jet flame under investigation in this paper can be accessed from Sandia TNF webpage. The flames are unpiloted and issue from 8.0-mm diameter nozzle, centred at the exit of a vertical wind tunnel, into a coflowing air stream of velocity 0.3m/s. Both the jet and coflow were recorded as being at ambient temperature at the burner exit plane. Flame contours and schematics of fuel and coflowing air stream locations shown in Fig. 1(a).

4.2 Prediction of flame structures

For the generation of flamelet data, it is assumed that the local balance between diffusion and reaction is similar to the one found in a prototype laminar flame. The control parameters of the flamelet are the mixture fraction, f and the strain rate, a . It is essential in the simulation that the mixture fraction field be adequately represented, because the mixture fraction statistics completely determine the thermochemical state of the flame. Fig.1 (b) represents the radial profiles of mean mixture fractions and its variances at six axial locations.

Comparisons between the predicted radial distributions of mixture fractions and its variances for both flamelet combustion models with differential diffusion (solid lines) and unity Lewis number flamelet model (dashed lines), along with the measurements at various near- and far-field axial stations. As can be seen from the figure, the predicted results show very little differences between the differential diffusion and unity Lewis number flamelet models and further results are presented only with the unity Lewis number assumptions. The overall agreements between the mixture fraction predictions and the measurements are reasonably good by the combustion models however, there are slight overpredictions found in the regions $2 < r < 10$ mm at $x/D=05$, $2 < r < 14$ mm at $x/D=10$, $2 < r < 20$ mm at $x/D=20$, $5 < r < 30$ mm at $x/D=40$. In the profiles of mixture fraction variances better predictions have been observed in the results obtained At $X/D=5$, the maximum variance found around the radial locations $3.5 \leq r \leq 6.2$ and overpredicted slightly by the models. At $X/D=10$, combustion models

overestimated the mixture fraction variance from the centreline towards the peak value and then good predictions are observed in the far axis region. At the locations $X/D=20$ and 40 , good predictions are observed with the exception of slightly overprediction if the peak values. At $X/D=60$ and 80 , the model predictions are well and shown the gradual decay of fluctuation in mixture fraction along the radial direction.

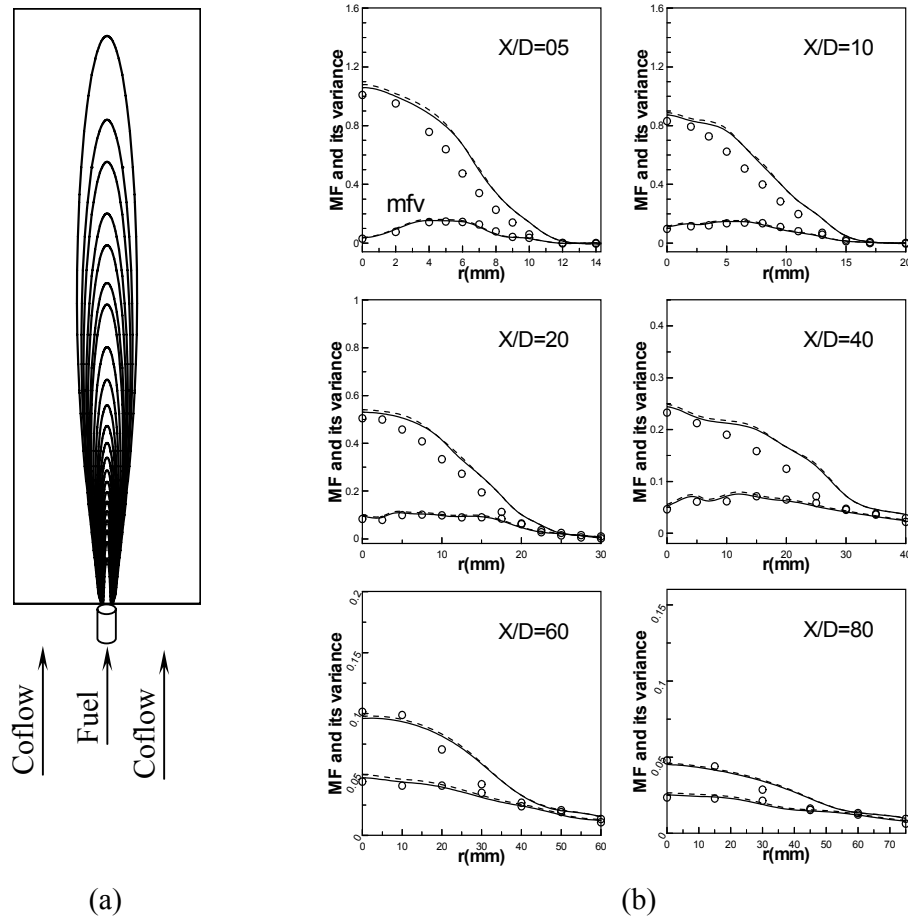


Figure 1: (a) Contours of flame structures with schematics of fuel and coflowing air stream locations. (b) Radial profiles of mean mixture fractions and its variances (mfv) at six axial locations: o measurements (Sandia, TNF); — flamelet model with differential diffusion; --- unity Lewis number flamelet model.

4.3 Prediction of flame with NOx

Predicted results of the mass fractions of NO, H₂O, H₂, O₂ and mean temperature distributions at different axial locations are compared with the measurements in Fig. 2. Predicted NO results compared as NO*500 with other species due to the production rate of NO is too small in comparison with the other species. Radial profiles of the mass fractions of H₂ and O₂ at six axial locations are similar to the mixture fraction profiles. The overall model predictions of H₂ are good with the exception of results in the near axis region where slight overpredictions are found for $X/D = 05$ to $X/D=40$. At $X/D = 60$ and 80 experimental data shows no level of H₂ but the model predictions have shown very little amount of existence of the Hydrogen. In the prediction of mass fraction of O₂ profiles at all axial locations combustion model have shown fairly good results with slight overpredictions. The profiles of O₂ are

better predicted by the model at the locations $X/D=05$ to 40 than that of at $X/D=60$ and 80 . The significant amount of overpredictions are found in the predictions of the temperature profiles by the combustion model. The peak temperatures predicted very near to the measured locations but maximum values are higher than those of measurements within the range of 200K to 500K . The significant over predictions in temperature profiles were also observed by Dally *et al.*(1998) and other authors as well. Dally *et al.* (1998) stated that the lower measured temperature compared with calculation is due to the averaging effects as a result of the intermittency in the flame at some locations and the overprediction is not due to the shortcomings in the simulation. The predicted temperature profiles have shown rapid decay from the maximum position similar to the experimental data. At $x/D=10, 20$ and 40 , the agreements between the measurements and the predictions are good with slight higher rate of overprediction by the model in the fuel rich zones than that of fuel lean zone. The predicted radial profiles of mass fraction of H_2O are in good agreement at all the axial locations with the experiment but slight overpredictions are found in the peak values and near axis region in some stations. The combustion model predicted the H_2O profile well in the far axis region from the peak value at $x/D=05$ to 60 . At $x/D=80$ overprediction is found between the centreline and $r=90\text{mm}$.

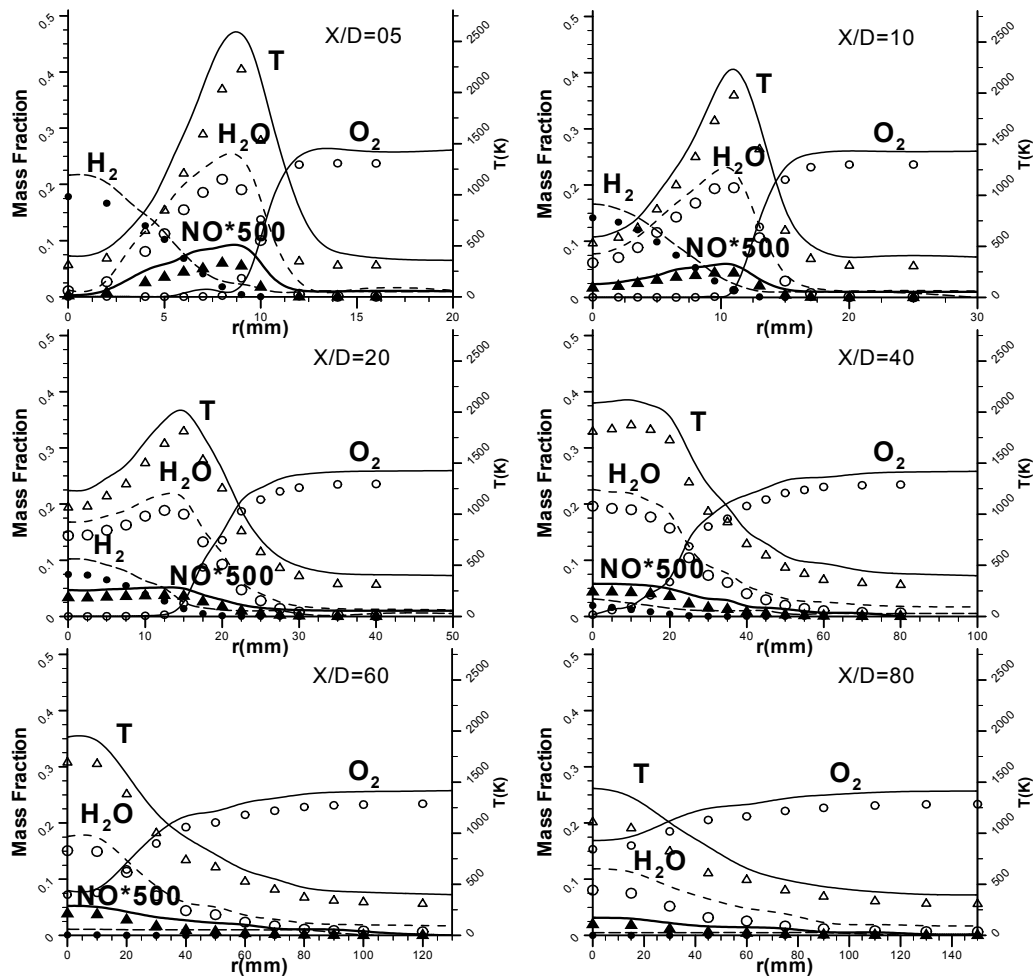


Figure 2: Predicted and measured mass fractions of NO , H_2O , H_2 , O_2 and mean temperature distributions: Computation (lines); measurements (symbols) [Sandia, TNF].

Comparisons of predicted NO mass fractions in Fig. 2 indicate that flamelet based NO_x modelling is able to predict the NO with fairly good level of accuracy. The small differences in the mixture fraction and scalar dissipation rate contribute to the slight overprediction of NO source terms and consequently the NO_x levels. The computed NO level at the location of lower flame temperatures also shows good agreement with the measurements. The maximum NO level observed in the hottest location the diffusion flame and it decaying gradually in the downstream area with decreasing temperature. At X/D = 05 and 10 the peaks NO found at the same radial position where temperature is maximum. The NO predictions and measurements show that the NO level is low in the rich part (near the axis) of the flame for X/D = 05 and 10. The NO level then reaches its peak further away from the axis near the flame front. Further downstream, the gradual decaying in the level of NO found with increasing radial distance. The results support the strong temperature dependence, and yield good agreement with measurements. Because the NO correlates well with the peak temperature in these types of flames.

5. Conclusions

Encouraging agreement between predictions with flamelet combustion model and the experimental data is demonstrated in this study. The flamelet combustion model provides an efficient method to model the H₂/N₂ jet diffusion flame. The predicted results of temperature and major and minor species as well as NO are compared against the reported experimental data. In the flamelet combustion model, the mean temperature, density and composition in the turbulent field are obtained by appropriately averaging the flamelets. On the other hand, the concentration of NO is calculated by solving its own transport equation with the source term obtained from the flamelet library. The effects of differential diffusion are also investigated and found very small which are shown in the mixture fraction results. Temperature and concentration of major and minor species and NO are also well reproduced with slight overprediction by the combustion model. Further improvement in the model predictions can be made in the several areas by constructing the flamelet libraries with the heat loss effects, using enthalpy defect or more accurate radiation model. With these future improvements, the flamelet combustion model is quite promising for evaluation of the influence of operating parameters in pollutant emissions.

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