CFD modelling of turbulent combustion and heat transfer

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CFD Modelling of Turbulent Combustion and Heat Transfer

by

Md Abdul Alim

A Doctoral Thesis

Submitted in partial fulfilment of the requirements for the award of
Doctor of Philosophy of Loughborough University

School of Mechanical and Manufacturing Engineering
Loughborough University

March 2004

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Abstract

This thesis is concerned with the development and implementation of computational fluid dynamics (CFD) based prediction methodologies for turbulent reacting flows with principal application to turbulent diffusion flame combustors. Numerical simulation of combustion problems involve strong coupling between chemistry, transport and fluid dynamics. The works accomplished in this study can be separated mainly into three distinct areas: i) assessment of the performance of turbulent combustion models and to implement suitable submodels for combustion and flame behaviour into CFD code; ii) Conducting CFD modelling of turbulent diffusion flames, radiation heat loss from combustion and flame zones; and iii) modelling of pollutants like NOx (oxides of nitrogen), identification of the effect of radiation heat loss on NOx formation.

The combustion models studied are the flame-sheet, equilibrium, eddy break-up and laminar flamelet models. An in-house CFD code is developed and combustion models are implemented. The basic numerical issues involving the discretisation schemes are addressed by employing three discretisation schemes namely, hybrid, power law and TVD (total variation diminishing) schemes.

The combustion of different fuels ranging from simple H2/N2 and CO/H2/N2 to complex CH4/H2 are investigated for different inlet velocities and boundary conditions. The performances of the combustion models are analysed for these fuels. The configurations used for the validation and assessment of the combustion models are co-flowing jet flames and bluff body burner stabilized flames. The high quality experimental databases available from Sandia national laboratories, the University of Sydney and other reported measurements are used for the purpose of evaluating the combustion models. The predicted results demonstrate the effects of turbulent mixing and the effects of chemical reactions on the combustion models.

The calculations show that all the combustion models like flame-sheet and equilibrium models are found to be inadequate even for the near equilibrium flames. Although the equilibrium chemistry model is capable of predicting the mixture fraction, temperature and concentrations of major and minor species, the predictive accuracy is found to be inadequate specially, when compared to the experimental data. In situations, where finite rate chemistry effects are important the laminar flamelet model is a good choice.
The importance of radiation heat loss in flamelet modelling of turbulent combustion has been demonstrated and a coupled flamelet/radiation modelling strategy is presented that combines the prediction of finite rate chemistry effects with a detailed solution of the radiative heat exchange. The coupling between the radiation and combustion is achieved through an additional parameter, the so-called enthalpy defect, which is defined as the difference between the actual enthalpy and the adiabatic enthalpy of a flame.

The present study demonstrates that the radiation adjusted flamelet model for combustion leads to encouraging results that reproduce the overall characteristics of radiating turbulent diffusion flames. Also the thermal radiation from flames reduces the local temperatures sufficiently to affect the production rate of NO and the incorporation of radiation heat transfer in combustion model is found to improve the prediction of the species NO significantly.

The key contributions of this thesis are as follows:

- Modification of in-house CFD code for turbulent reacting flow and development of CFD based iterative scheme for the turbulent diffusion flames to account for radiation heat loss from combustion and flame zones.

- Thorough assessment of turbulent combustion modelling techniques for different cases of diffusion flames, demonstration of the importance of differential diffusion in the flamelet modelling of combustion and comprehensive validation.

- Demonstration of the importance of radiation heat loss in the modelling of turbulent combustion, implementation of radiation modelling in the three cases of diffusion flames and comprehensive validation of CFD based combustion radiation results.

- Development of modelling strategy for the pollutants like oxides of nitrogen (NOx), implementation of NOx modelling in the different flames cases and identified the effect of radiation heat loss on NOx formation.

The works addressed in this thesis are presented with the applications to turbulent diffusion flame combustors. However, these works can easily be extended to the industrial applications and applied to a large variety of other challenging domains.
Acknowledgements

I would especially like to thank my supervisor, Dr. W. Malalasekera, for indoctrinating me into the ways of science, computer simulations, combustion modelling, and turbulence, for his steadfast support, limitless patience, and encouragement that he has given to me. I also like to express my deep gratitude and indebtedness to him for his supervision, guidance, invaluable suggestions and constructive criticism throughout this investigation. Thanks to Professor Colin Garner for his suggestions, comments, valuable discussions and encouragement in the research work.

This work has benefited greatly from the suggestions of Professor A. Masri who was a visitor from Sydney University. I would like to thank him for providing valuable new experimental data sets.

The financial support and other assistance provided by the School of Mechanical and Manufacturing Engineering, Loughborough University is gratefully acknowledged.

I wish to acknowledge the friendly help extended by Dr. Mamdud Hossain from the very beginning of this study and providing me the computer codes he used before including RUN-1DL and guiding me towards using those.

I save the last special thanks for my wife, Azmun Nahar Nipa, she stood by me through the most difficult times of this process, offering her unconditional love, support and patience. You are an inspiration indeed, thank you.
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Appendix A

Appendix B

References
Nomenclature

Roman Letters

\(a\)  
strain rate [\(\text{s}^{-1}\)]

\(a\)  
exponent in beta pdf, Eq.(3.9)

\(a_B\)  
temperature exponent in forward and backward reaction rate, Eq.(2.25)

\(a_p, a_E, a_W, a_N, a_s\)  
coefficients of discretised equation, Eq. (3.2)

\(A\)  
area [\(\text{m}^2\)]

\(A\)  
model constant in Magnussen's EBU model, Eq. (2.88)

\(b\)  
exponent in beta pdf, Eq. (3.9)

\(B\)  
model constant in Magnussen's EBU model, Eq. (2.88)

\(B_\beta\)  
pre-exponential factor, Eq.(2.25)

\(C_{EBU}\)  
model constant in EBU model, Eq. (2.87)

\(C_{\varepsilon_1}, C_{\varepsilon_2}\)  
turbulence model constants in the \(\varepsilon\)-equation, Eq. (2.40)

\(C_\mu\)  
model constant in the definition of turbulent viscosity, Eq. (2.38)

\(C_{g_1}, C_{g_2}\)  
constants in transport equation of mixture fraction variance, Eq. (2.33)

\(C_p\)  
mixture specific heat at constant pressure [\(\text{J/kg K}\)]

\(C_x\)  
proportionality constant, Eq. (2.50)

\(d\)  
diameter of fuel jet nozzle, diameter of inlet pipe [\(\text{m}\)]

\(D\)  
internal diameter of wind tunnel, diameter of bluff body [\(\text{m}\)]

\(D_{T,i}\)  
thermal diffusion coefficient of species \(i\)

\(D_\alpha\)  
diffusion coefficient of species \(\alpha\) [\(\text{m}^2/\text{s}\)]

\(E\)  
constant in log-law wall function Eq. (3.8)
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tr>
<td>$E_{i\beta}$</td>
<td>activation energy, Eq. (2.25) [J/mol]</td>
</tr>
<tr>
<td>$E_g$</td>
<td>black body emissive power [W/m$^2$]</td>
</tr>
<tr>
<td>$\tilde{f}$</td>
<td>nondimensional velocity profiles, Eq. (2.71)</td>
</tr>
<tr>
<td>$g_i$</td>
<td>gravitational acceleration vector</td>
</tr>
<tr>
<td>$G$</td>
<td>production rate of kinetic energy, Eq. (2.41)</td>
</tr>
<tr>
<td>$h$</td>
<td>enthalpy of mixture [J/kg]</td>
</tr>
<tr>
<td>$H_{fu}$</td>
<td>heating value of fuel [J/kg]</td>
</tr>
<tr>
<td>$i$</td>
<td>turbulence intensity</td>
</tr>
<tr>
<td>$I$</td>
<td>radiation intensity</td>
</tr>
<tr>
<td>$J^k_i$</td>
<td>molecular diffusive flux of the species $k$ for $i$th coordinate.</td>
</tr>
<tr>
<td>$K$</td>
<td>turbulence kinetic energy [m$^2$/s$^2$]</td>
</tr>
<tr>
<td>$k_{i\beta}$</td>
<td>Arrhenius rate coefficient of forward reaction, Eq. (2.24) [/s]</td>
</tr>
<tr>
<td>$k_{\beta i}$</td>
<td>Arrhenius rate coefficient of backward reaction, Eq. (2.24) [/s]</td>
</tr>
<tr>
<td>$k_g$</td>
<td>gas absorption coefficient</td>
</tr>
<tr>
<td>$L$</td>
<td>radiation path length, [m]</td>
</tr>
<tr>
<td>$l_m$</td>
<td>turbulent mixing length, Eq. (3.7)</td>
</tr>
<tr>
<td>$l_{\beta}$</td>
<td>order of backward reactions, Eq. (2.24)</td>
</tr>
<tr>
<td>$m_{\beta}$</td>
<td>order of forward reactions, Eq. (2.24)</td>
</tr>
<tr>
<td>$M'_{\alpha}$</td>
<td>chemical symbol of species $\alpha$, Eq. (2.22)</td>
</tr>
<tr>
<td>$M_{\alpha}$</td>
<td>molar mass or molecular weight of species $\alpha$, [kg/mol]</td>
</tr>
<tr>
<td>$M_{in}$</td>
<td>mass flow rate at inlet [kg/s]</td>
</tr>
<tr>
<td>$M_{out}$</td>
<td>mass flow rate at outlet [kg/s]</td>
</tr>
<tr>
<td>$p, P$</td>
<td>pressure [N/m$^2$]</td>
</tr>
<tr>
<td>$P\tilde{O}$</td>
<td>probability density function</td>
</tr>
<tr>
<td>$P_c$</td>
<td>critical probability of extinction</td>
</tr>
<tr>
<td>$P_{ext}$</td>
<td>probability of extinction</td>
</tr>
<tr>
<td>$P_e$</td>
<td>Peclet number</td>
</tr>
<tr>
<td>$q_R$</td>
<td>radiative heat flux</td>
</tr>
<tr>
<td>$q_w$</td>
<td>wall heat flux due to radiation [W/m$^2$]</td>
</tr>
</tbody>
</table>
Nomenclature

- $r$: radius of inlet pipe
- $\hat{R}$: Universal gas constant = 8.3143 [J/mol]
- $Re_t$: turbulence Reynolds number
- $s$: stoichiometric mass of oxygen [kg]
- $S$: strain-rate tensor, $S_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i})$
- $S_h$: radiation source term, [W/m$^3$]
- $S_\phi$: general source term in transport equations, Eq. (3.1)
- $S_c, S_p$: general source term in discretised equation, Eq. (3.3)
- $S_c$: Schmidt number
- $t$: time [s]
- $t_k$: Kolmogorov time [s]
- $t_c$: chemical time [s]
- $T$: temperature [K]
- $T_g$: gas temperature [K]
- $u, U$: axial velocity [m/s]
- $u_i$: Cartesian velocity components
- $v, V$: radial velocity [m/s]
- $V$: volume of computational cell [m$^3$]
- $V$: dimensionless velocity, Eq. (2.40)
- $x$: axial distance [m]
- $x_i, x_j$: Cartesian coordinates
- $x_i$: mole fraction of species $i$
- $y$: radial distance [m]
- $Y_i, y_i$: mass fraction of species $i$
- $Z$: mixture fraction
- $Z_m$: mass fraction of element $m$
- $Z_{st}$: stoichiometric mixture fraction
- $Z^2$: mixture fraction variance
Greek Symbols

\[ \Gamma \]  \hspace{1cm} \text{diffusive transport coefficient} \\
\[ \Gamma() \]  \hspace{1cm} \text{Gamma function, } \Gamma(n) = (n-1)! \\
\[ \delta() \]  \hspace{1cm} \text{delta function} \\
\[ \delta_{ij} \]  \hspace{1cm} \text{Kronecker delta} \\
\[ \Delta Z \]  \hspace{1cm} \text{reaction zone thickness in mixture fraction space} \\
\[ \varepsilon \]  \hspace{1cm} \text{rate of turbulent kinetic energy dissipation} [m^2/s^3] \\
\[ \varepsilon_g \]  \hspace{1cm} \text{emissivity of gas} \\
\[ \zeta \]  \hspace{1cm} \text{enthalpy defect, } \Delta h \\
\[ \eta \]  \hspace{1cm} \text{non dimensional distance, Eq. (2.73)} \\
\[ \kappa \]  \hspace{1cm} \text{von Karman constant in log-law wall function, Eq. (3.8)} \\
\[ \mu \]  \hspace{1cm} \text{molecular viscosity of mixture} [kg/m s] \\
\[ \mu \]  \hspace{1cm} \text{mean value of log-normal pdf, Eq. (3.20)} \\
\[ \mu_t \]  \hspace{1cm} \text{turbulent viscosity} [kg/m s] \\
\[ \nu_{\alpha \beta}, \nu_{\alpha \beta}^* \]  \hspace{1cm} \text{stoichiometric coefficients of species } \alpha \text{ in reaction step } \beta, \text{ Eq. (2.22)} \\
\[ \rho \]  \hspace{1cm} \text{density} [kg/m^3] \\
\[ \sigma \]  \hspace{1cm} \text{Prandtl number} \\
\[ \sigma \]  \hspace{1cm} \text{standard deviation of log-normal pdf, Eq. (3.20)} \\
\[ \sigma_t \]  \hspace{1cm} \text{turbulent Prandtl number} \\
\[ \sigma_k \]  \hspace{1cm} \text{Prandtl number governing diffusion of turbulent kinetic energy, } k \\
\[ \sigma_e \]  \hspace{1cm} \text{Prandtl number governing diffusion of turbulent energy dissipation rate, } \varepsilon \\
\[ \tau_{ij} \]  \hspace{1cm} \text{viscous stress tensor, Eq. (2.16)} [N/m^2] \\
\[ \tau_w \]  \hspace{1cm} \text{wall shear stress} [N/m^2] \\
\[ \phi \]  \hspace{1cm} \text{scalar variable} \\
\[ \chi \]  \hspace{1cm} \text{scalar dissipation rate} [/s] \\
\[ \chi_a \]  \hspace{1cm} \text{scalar dissipation rate at the extinction limit} [/s] \\
\[ \psi \]  \hspace{1cm} \text{limiter in TVD scheme, Table (3.1)} \\
\[ \Omega \]  \hspace{1cm} \text{solid angle}
\dot{\omega}_\alpha \quad \text{net rate of formation of species } \alpha \ [\text{mol/m}^3 \text{ s}]

**Subscripts and Superscripts**

- \(\alpha\) \hspace{1cm} \text{species}
- \(A\) \hspace{1cm} \text{air}
- \(b\) \hspace{1cm} \text{black body}
- \(C\) \hspace{1cm} \text{carbon}
- \(DC\) \hspace{1cm} \text{deferred correction}
- \(eq\) \hspace{1cm} \text{equilibrium}
- \(F, f, fu\) \hspace{1cm} \text{fuel}
- \(g\) \hspace{1cm} \text{gas}
- \(H\) \hspace{1cm} \text{hydrogen}
- \(in\) \hspace{1cm} \text{inlet}
- \(NO\) \hspace{1cm} \text{nitric oxide}
- \(ox\) \hspace{1cm} \text{oxygen}
- \(out\) \hspace{1cm} \text{outlet}
- \(O\) \hspace{1cm} \text{oxygen}
- \(pr\) \hspace{1cm} \text{product}
- \(st\) \hspace{1cm} \text{stoichiometric}

**Diacritical Marks**

- \(\bar{\phi}\) \hspace{1cm} \text{time-averaged mean quantity of } \phi
- \(\tilde{\phi}\) \hspace{1cm} \text{Favre-averaged mean quantity of } \phi
- \(\phi'\) \hspace{1cm} \text{turbulent fluctuation of } \phi \text{ after time-averaging}
- \(\phi''\) \hspace{1cm} \text{turbulent fluctuation of } \phi \text{ after Favre-averaging}
# Abbreviations

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tbody>
<tr>
<td>CDS</td>
<td>central differencing scheme</td>
</tr>
<tr>
<td>CFD</td>
<td>computational fluid dynamics</td>
</tr>
<tr>
<td>CMC</td>
<td>conditional moment closure</td>
</tr>
<tr>
<td>CPU</td>
<td>central processing unit</td>
</tr>
<tr>
<td>DNS</td>
<td>direct numerical simulation</td>
</tr>
<tr>
<td>DTM</td>
<td>discrete transfer method</td>
</tr>
<tr>
<td>EBU</td>
<td>eddy break-up</td>
</tr>
<tr>
<td>EDC</td>
<td>eddy dissipation concept</td>
</tr>
<tr>
<td>IEM</td>
<td>interaction by exchange with the mean</td>
</tr>
<tr>
<td>ILDM</td>
<td>intrinsic low-dimensional manifold</td>
</tr>
<tr>
<td>LIF</td>
<td>laser induced fluorescence</td>
</tr>
<tr>
<td>pdf</td>
<td>probability density function</td>
</tr>
<tr>
<td>QUICK</td>
<td>quadratic upstream interpolation for convection kinematics</td>
</tr>
<tr>
<td>rms</td>
<td>root mean square</td>
</tr>
<tr>
<td>RSM</td>
<td>reynolds stress model</td>
</tr>
<tr>
<td>SIMPLE</td>
<td>semi-implicit method for pressure linked equations</td>
</tr>
<tr>
<td>TDMA</td>
<td>tri-diagonal matrix algorithm</td>
</tr>
<tr>
<td>TNF</td>
<td>turbulent nonpremixed flame</td>
</tr>
<tr>
<td>UMIST</td>
<td>upstream monotonic interpolation for scalar transport</td>
</tr>
<tr>
<td>TVD</td>
<td>total variation diminishing</td>
</tr>
<tr>
<td>WSGG</td>
<td>weighted-sum-of-gray-gases</td>
</tr>
</tbody>
</table>
Introduction

1.1 Motivation and Objectives

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. Therefore it is really worthwhile studying the process of combustion.

Combustion is the rapid chemical reaction between substances that is usually accompanied by generation of heat and light in the form of flame. In all cases, oxygen comprises one of the reactants. Other physical phenomena that sometimes occur during combustion reactions are explosion and detonation. Combustion, one of the most important classes of chemical reaction, is often considered as a point of greatest intensity, in the oxidation of certain types of substances. Although most flames have regions where reduction reactions are important, combustion is primarily the combining of combustible material with oxygen. Flame is the Rapidly reacting body of gas, commonly a mixture of air and a combustible gas, that gives off heat and, usually, light and is self-propagating. Mixtures of natural gas and air, for example, will not propagate
flame if the proportion of gas is less than about 4 percent or more than about 15 percent \textit{(Encyclopaedia Britannica)}.

Because of the critical role of combustion the accurate control of turbulent flames appears as a real challenge in the production of power, a great deal of research effort is devoted to finding ways to burn fuels more efficiently. Objectives of many research is to seek how to reduce the pollutants released by combustion, since these products contribute greatly to environmental problems. CO$_2$ is recognised as one of the contributors to the greenhouse effects and NOx is one of the precursors of the acid rain. Detailed knowledge and a better understanding of different processes involved in the turbulent combustion are therefore essential for the dual purpose of increasing combustion efficiency and reducing pollutant emissions. In many combustion laboratories sophisticated laser probes are also used to study boilers and engine systems in operation, in order to discover sites of fuel wastage and to improve the design of these fuel-burning systems. Lasers can also be used to probe the chemical kinetics of flames themselves, in order to understand better the ways and uses of fire.

Continuing demands are there to increase efficiency of gas turbines, furnaces, aero engines and industrial combustors and a conflicting concern over the problems related to environment pollution and noise. All these have, in the past few decades, resulted in a tremendous burst of interest and research activities in the field of combustion. The simultaneous occurrence of chemical reaction and transport of mass, momentum and energy makes analysis of these problems extremely difficult. Moreover, practical flames are mostly turbulent because of the requirement to produce a high volumetric rate of energy for efficiency and compactness. This complicates the combustion research further even if one addresses the problem at a simplified level in which practical difficulties might be neglected.

\textit{Radiative heat transfer} also plays an important role and being the dominant mode of heat transfer in fires and in many high temperature combustion systems. In furnaces, walls also interact with combustion through radiative heat transfer. Advances in measurement techniques and the application of laser diagnostics systems (Dibble et al. 1987) to carefully conceived turbulent flames have greatly advanced the understanding of the finite rate chemistry effects in turbulent flames. Detailed measurements of turbulent flames have been reported, including the scatter plots of major and minor species for a number of fuels and burner configurations (Masri et al. 1988,
Dally et al. (1996,1998) have provided extensive single point measurements of reactive and conserved scalars in a range of bluff body stabilised turbulent non-premixed flames. These measurements have revealed extensive and novel information about local structures of turbulent flames. Local extinction and bimodality of measured probability density functions of some reactive scalars are observed leading to blow-off of the flames (Masri et al. 1996). Masri et al. (1994) have shown that the finite rate chemistry effects are not important inside the recirculation zone even when the flame exhibit local extinction at the neck zone of the bluff body flame. Masri et al. (1996) have also shown that the flames are not strictly at equilibrium conditions even when the flame is far away from blow-off. The hydroxyl radical exists in super-equilibrium quantity and the temperature and mass fraction of major species also deviate, though slightly, from the equilibrium condition even at longer residence time. The presence of instantaneous concentrations of some species in excess of the steady laminar flamelet limits have also been reported (Masri et al. 1996). These findings have encouraged researchers to revisit such flows with both experimental and numerical investigations.

Computing is now truly on par with experiment and theory as a research tool to produce multi-scale information that is not available by using any other technique. Computational fluid dynamics (CFD) is efficiently used to improve the design of systems, and today no real progress in design can be made without using CFD.

Turbulent combustion research is also driven by the demand for the development of adequate predictive tools for the design and analysis of practical combustion situations which are encountered in industrial applications. In the modelling, a number of simplifying assumptions are made in the representation of the physical and chemical processes. An essential part of the development of models is the validation of these assumptions. The validation is generally done by comparing the model calculations against the detailed experimental data. From the practical point of view, the experimental configuration used for validation purposes should be simple and well defined, so that the boundary conditions can be set without any ambiguity in the computation. The experimental configuration should also allow the use of the modern non-intrusive laser diagnostic techniques. These requirements make industrial combustors unsuitable for detailed validation studies. Simple jet flames, piloted jet flames and bluff body stabilised flames are generally used as benchmark cases for the
comparison of model predictions. These validation studies are not only to identify the model that best matches the experimental data, since a model can give a right result for wrong reasons, but also to distinguish the differences in model calculations that result from applying different submodels. Because of the large number of degrees of freedom involved in turbulent combustion, a full simulation of a practical system cannot be performed and averaging techniques leading to unclosed equations are necessary. *Models for turbulent flames* are then developed: closure techniques are proposed for unknown terms found in exact averaged balance equations. Once the models have been implemented in numerical codes, *validation procedures* are required. The numerical modeling is validated against measurements obtained from experiments. Configurations as close as possible to actual industrial systems are chosen for these tests. Then, the ultimate step is the simulation of a real combustion device.

The problem of particulate formation during incomplete combustion of hydrocarbons is an issue of practical importance. It demands a molecular-level understanding of the processes forming soot and products of high molecular weight to develop strategies to control emissions. Most of the modelling of hydrocarbon growth has been performed in zero-dimensional and one-dimensional geometries, such as in shock-tubes and premixed flames to uncouple the complex chemistry from the fluid dynamics. Indeed, to describe for practical combustion systems the fluid dynamics with turbulence and the detailed chemistry, even for very simple hydrocarbons, would require tens of chemical species and hundred of reactions, with the simultaneous calculation of a very large number of dependent variables. The main objective of such a detailed kinetic mechanism is to simulate the pyrolysis and oxidation of hydrocarbons over a wide range of operating conditions, particularly in real devices. It is therefore important that the reliability of a kinetic scheme be tested over the widest possible range of temperature, C/O ratio and pressure. In diffusion flames, there are changes from room temperature up to the flame temperature and the local C/O ratio spans the range from the fuel side (C/O = ∞) to the air side (C/O = 0).

Modelling of diffusion flames initially focused on predicting concentration profiles of major species (Rogg et al. 1986, Ramakrishna et al. 1995). A few studies gave particular attention to improvements in the quantitative prediction of key-species in soot formation, such as acetylene and benzene (Rogg et al. 1986) and in modelling soot
formation using detailed gas-phase chemistry (Yamaoka and Tsuji 1974) and
(Ramakrishna et al. 1995). The agreement with experiments had generally been
acceptable; however, quantitative predictions of particulates remained limited by the
ability of the kinetic mechanisms to predict accurately the concentrations of important
precursors, particularly aromatics of high molecular mass on the fuel side of the flame.
A kinetic mechanism able to predict the formation of the heavy aromatics in shock-
tubes, rich pre-mixed flames and opposed-flow diffusion flames has also been

To predict flame properties, such as transport and chemical kinetics, burning velocities,
extinction limits, flammability limits, radiation heat loss and its effect on species
production. This study focuses primarily on chemical kinetics, turbulence, radiation
heat transfer, production of pollutant like CO and NOx and their control. To be able to
control pollution, we must first understand how pollutants are produced in combustion
processes. This understanding requires much basic chemical information. Combustion
chemistry theory and modeling techniques provide such information. For example,
 electronic structure theory provides the detailed potential energy surfaces of elementary
chemical reactions and the thermochemistry of unstable free radicals. Dynamics theory
uses these potential energy surfaces to yield rates and product distributions of the
elementary reactions. All of this information are used to develop chemical kinetic
models of combustion, which then used to simulate macroscopic experiments and
industrial processes. Current practice in the industry relies heavily on RANS
calculations, backed up by expensive physical testing. The trend in the industry and
engineering in general is towards shorter design cycles through increased reliance on
numerical prediction. RANS models for turbulent combustion do not appear capable of
meeting the needs of industry in this regard, and the industry is beginning to look
towards more sophisticated prediction tools that can take advantage of new
computational capabilities. It is, in fact, the relentless advance of computer technology
that is the driving force behind increasing expectations for computational fluid
dynamics and the main motivation for this study.

The objectives of the study are to develop and implement CFD based prediction
methodologies for turbulent reacting flows with principal application to turbulent
diffusion flame combustors. In the industrial application where accurate high fidelity prediction methods for turbulent combustion are desperately needed for the design of next generation, low emissions combustors.

The main targets of this study are to assess the performance of the turbulent combustion models and to implement suitable submodels for combustion and flame behaviour into CFD code; Using CFD to investigate the behaviour of diffusion flames. Reviewing/implementing computationally advanced combustion and turbulence models into CFD code. Conducting CFD modelling of turbulent diffusion flames, radiation heat loss from combustion and flame zones, effect of radiation on NOx formation and validation of model predictions against experimental data.

1.2 Literature Survey

Turbulent combustion is a complex physico-chemical phenomenon which is spatially three-dimensional and is of transient nature. This phenomenon has been the subject of intense research over the past sixty years and continues to be of high priority in view of the worldwide concern about energy and pollution control (Givi 1989; Pope 1990; Libby & Williams 1994; Vervisch & Poinsot 1998). Turbulent flows with 'non-premixed' reactants are in use in the majority of practical combustion systems. Examples are, to name a few, gas and oil furnaces and burners, diesel engines, and hypersonic propulsion systems.

The transport and chemical structure of flames are the obvious subjects for combustion studies, and yet the study of flames has always posed experimental difficulties. Moreover, during the last few decades, there are continuing demands for increased efficiency for gas turbines, furnaces, aero engines and other combustors and a conflicting concern over the problems related to environment pollution and noise. There are many types of flames, for example turbulent and laminar flames or diffusion and premixed flames. The trick to understanding flames is to know the chemical kinetics and the flow well enough to model the interaction between the two. Laser and mass-spectrometer-based diagnostics and modern computational tools give hope that we can investigate model systems in great detail.
The governing transport equations are the well known conservation equations of mass, momentum, energy and species concentrations. The transport mechanism and the thermodynamic properties of the species involved in a typical combustion situation are well established. Furthermore, the detailed reaction mechanisms of the gaseous fuels involved in practical applications are proven to be reliable and accurate. Despite all these, the crucial problem lies in the solution of governing equations for the turbulent combustion under investigation. The physical processes of convection, diffusion and reaction in turbulent flames involve a wide range of length and time scales. The computational efforts needed to resolve the length and time scales by the direct numerical solution method exceed, by a large margin, the capacity of present day computers for virtually all turbulent combustion situations of practical interest. The complete deterministic treatment of the governing equations for the practical combustion situation is not possible in the foreseeable future. However, this has been a major driving force behind many research efforts for the development of simplified yet physically realistic descriptions of turbulent combustion.

The present study focuses on the modelling of turbulent non-premixed combustion. In this type of combustion, the fuel and air are fed separately into the combustor where chemical reaction takes place. This is in contrary to the premixed flame, where fuel and air mix before reaching the combustion chamber. For safety reasons, combustion in most of the industrial furnaces and gas turbines are of the non-premixed type.

There are currently four main methods of turbulent combustion modelling approaches available for incorporation into CFD: (a) conserved scalar, (b) eddy beak-up, (c) laminar flamelet (d) pdf transport/Monte-Carlo method. The last method is theoretically the most accurate and is capable of handling the reaction rate terms without any modelling assumptions. However, the pdf transport/Monte-Carlo model is very resource-intensive and the application of the model for the industrial calculations is still not wide spread. The remaining three models are viable options for industrial applications. Other combustion modelling approaches available to the user include the conditional moment closure (CMC) and interaction by exchange with the mean (IEM) model, which are, however, not widely used in the practical situations.
The conserved scalar method is the simplest of all models and was first devised in the early 70's. The central assumption of the model is that, whenever fuel and oxygen exist at a point the chemical reaction will instantaneously proceed to completion in a single step reaction, producing combustion products (Elghobashi and Pun 1974; Elghobashi 1979). The ensuing simple chemical reaction system (SCRS) relates the thermo-chemical state of a flame with a conserved scalar, known as the mixture fraction through algebraic relations. The main problem of determining the chemical reaction rate is thus reduced to determining the mixture fraction and its variance. The "brush" like thick reaction zone, often the concern in early research, was determined through incorporating the influence of turbulent fluctuations. Following the work of Spalding (1971a), who first demonstrated the incorporation of the calculations of turbulent fluctuations in a turbulent jet issuing into stagnant surroundings, the incorporation of the turbulent fluctuations into combustion modelling through a probability density function (pdf) was demonstrated by Elghobashi and Pun (1974), Elghobashi (1979), Lockwood and Naguib (1975). An alternative relation between the thermo-chemical state and the mixture fraction through chemical equilibrium formed the basis for determining the minor species CO and pollutants NO (Jones and Priddin 1978; Jones 1980). Though the equilibrium model was successful in predicting temperature and major species, it failed to predict the correct level of CO in hydrocarbon fuels. The equilibrium model also underpredicted the NO level by a wide margin. Jones (1980) speculated that the probability density function may be a source of errors, but according to him, a more widely accepted explanation is the equilibrium assumptions of atom O. The O atom can be an order of magnitude higher than the equilibrium values Jones (1980). Despite the shortcomings, the conserved scalar model has been widely used for a number of applications: furnace and gas turbine simulations (Smith and Smoot 1981; Shyy et al. 1988; Nikjoooy et al. 1988), finite element based combustion simulations (Kim and Chung 1987; Elkaim et al. 1993), three dimensional jet diffusion flames in a surface layer (Hernández et al. 1995), study of the effects of turbulence model on combustion (Gran et al. 1997), combustion including radiation heat transfer (Abbas et al. 1984; Ramamurthy et al. 1994) and the flow structure in bluff body flames (Dally et al. 1998) to mention a few.
In the eddy break-up (EBU) model, the problematic reaction rate term in turbulent combustion is modelled by considering the hydrodynamic features rather than the chemical kinetic features. The model was first formulated by (Spalding 1971b) with some modifications later (Spalding 1976). However, the most popular of the eddy-break models is the one proposed by Magnussen and Hjertager (1976). In the eddy break-up model, the reaction rate of the fuel is given by the rate of dissipation of the eddies containing the species of fuel, oxidiser and the products. The eddy break-up model of Magnussen and Hjertager (1976) differs from the original model of (Spalding 1971b) in relating the dissipation of the eddies to the mean concentrations instead of the concentration fluctuations. The EBU model has been shown to make reasonable predictions in a number of applications (Khalil et al. 1975; Magnussen and Hjertager 1976). In order to retain finite rate effects, Nikjooy et al. (1988) combined the eddy break-up expression with the Arrhenius type expression. Though the model looks promising, it lacks a sound theoretical foundation. An advanced eddy break-up model, commonly known as the eddy dissipation concept (EDC) model, treats the small scale eddies where the reaction takes place, as a perfectly stirred reactor (Gran 1994). This facilitates the incorporation of the finite rate effects in the combustion calculations. The importance of the finite rate effects in the EDC model is demonstrated by Gran and Magnussen (1996b). Their work shows that the fast chemistry EDC model is inferior to the conserved scalar model, but the finite rate EDC model improves the prediction considerably. The popularity of the EBU/EDC model lies in the fact that the model is simple and straightforward to implement. Bresslof et al. (1996) have shown an application of the EBU model where the combustion is treated with the simple EBU model while the complex issues of soot and radiation modelling are described in great detail.

The laminar flamelet model is a very powerful model, which includes the finite rate chemistry effects in the turbulent non-premixed combustion calculations without increasing the computational load considerably. From the application point of view, the laminar flamelet model uses the same computational framework as the conserved scalar model. The instantaneous relationship between the mixture fraction and all the scalar variables, such as the species mass fractions and the temperature, is computed from the laminar diffusion flame. The realistic reaction mechanism and the transport of species
can be included in the flamelet model as the flamelet profiles are calculated off-line. The mathematical background and the formulation of the laminar flamelet model can be found in Bray and Peters (1994), Peters (1984, 1986). The improvement in prediction by the laminar flamelet model over the equilibrium model was first demonstrated by Liew et al. (1984). Their calculation successfully predicted the presence of $O_2$ at the axis of the fuel jet which results from non-equilibrium effects. However, the prediction of CO was somewhat deteriorated. Drake et al. (1988) have shown that the flamelet model produces good predictions of temperature, mass fractions of $CO_2$, CO, $H_2O$ for a syngas jet flame. Their calculations have predicted the super-equilibrium OH level in the near field of the jet, but also predicted a rapid decay of OH towards equilibrium at the downstream location of the jet. Haworth et al. (1988) have shown that the history effect is important for the prediction of OH and using an equivalent steady strain rate to include the transient effect, they have predicted accurate levels of OH at the downstream location for the same syngas jet flame. The calculations of Lentini (1994) however contradict the findings of both Drake et al. (1988) and Haworth et al. (1988) for OH prediction. Lentini (1994) using the steady flamelet model has shown that the OH prediction in the upstream level is comparable to the predictions of Drake et al. (1988) and Haworth et al. (1988). More importantly, the prediction does not show a rapid decay towards the equilibrium level at the downstream location as observed by Drake et al. (1988). The laminar flamelet model is shown to be successful in predicting the complex chloromethane fuel combustion (Lentini and Puri 1995). The flamelet model has also been used for the prediction of a number of various practical issues such as NO formation (Sandars et al. 1997), soot (Brookes and Moss 1999), lift-off phenomenon (Sanders and Lamers 1994), partial premixing (Rogg et al. 1986) and transient effects (Pitsch et al. 1998) with varying degrees of success.

The pdf transport/Monte-Carlo model is the most promising combustion model for incorporating finite rate chemistry effects. The chemical reaction term is treated exactly without any modelling, but the model suffers from the modelling of the mixing term. The pdf transport/Monte-Carlo model is very resource-intensive. To minimise computational burden, often the reduced mechanism, ILDM (intrinsic low-dimensional manifold) mechanism, partial equilibrium and flamelet models are used to describe the thermo-chemical state instead of the full reaction mechanisms. A detailed description of
the pdf transfer/Monte-Carlo formulation can be found in Pope (1985, 1990). Because of the computational resources required, this model is still not viable for industrial applications.

The conditional moment closure (CMC) model developed by Bilger (1993) has shown good results for including finite-rate chemistry effects. The CMC model has been successful in predicting the emission of NO (Smith et al. 1992; Barlow et al. 1999). An assessment of the flamelet and CMC models against direct numerical simulation (DNS) data has shown the superiority of the CMC model (Swaminathan and Bilger 1999). Borghi's group (Obounou et al. 1994) have developed a finite rate chemistry model where diffusion is modelled by using the interaction by exchange with the mean (IEM) and the reaction rate term includes a time delay which is compared with a mixing time scale. The IEM model has been successful in a number of applications including the prediction of the scaling behaviour of NO (Schlatter et al. 1996). These new modelling approaches are still in their early stages of development and have not yet gained popularity.

Filtering and Large eddy simulation (LES) has been developed and studied as a turbulent flow prediction tool for engineering during the past three decades but the significant progress is occurring more recently with advances in computer technology and the development of the dynamic subgrid-scale modeling procedure (Germano et al. 1991). The objective of LES is to explicitly compute the largest structures of the flow (typically the structures larger than the computational mesh size), while the effects of the smaller one are modelled. With the dynamic procedure, model coefficients are automatically computed using information contained in the resolved turbulence scales, thereby eliminating the uncertainties associated with tuneable model parameters. Moin et al. (1991) applied the dynamic procedure to scalar transport and subgrid kinetic energy models for compressible turbulent flows using Favre filtering. Reviews of LES are given by Lesieur and Métais (1996) and Moin (1997). The application of large eddy simulation to chemically reacting flows has been a subject of growing interest, but to date few simulations of realistic combustion systems have been undertaken.

Turbulent combustion modelling is actually a continuous ring between theoretical studies to analyze combustion, understand flames and improve models, implementation of these models into CFD, experimental measurements and comparison between these experimental data and the numerical results.
The use of modern laser based diagnostic systems for combustion measurements has resulted in a number of well documented experimental data available through the worldwide web which are suitable as benchmark problems (Masri 1997; Barlow and Frank 1997; Hassel 1997). This in turn resulted in renewed interest in the validation studies of combustion models. The International Workshop on Measurement and Computation of Turbulent Nonpremixed Flames (TNF) are solely dedicated to different issues related to combustion modelling with special emphasis on the turbulence-chemistry interaction. The comparison of the combustion model calculations with a set of well documented experimental data made available through the simultaneous applications of Raman scattering, Rayleigh scattering and laser induced fluorescence (LIF) is essential in identifying the strengths or weaknesses of the combustion models under different situations. Validation studies are also necessary for further development of combustion models. In this regard it is important to isolate different submodels that are involved with the overall combustion calculations. To accomplish this, it is necessary to understand the differences in model predictions that result from using different combustion models, different fluid dynamics models, different model constants, different numerical schemes, different thermo-fluid properties and different radiation models. The emphasis of the present study is not only to identify a suitable model that fares well under different conditions but also to identify the influences of different submodels.

The combustion models evaluated here are the conserved scalar with flame sheet and equilibrium chemistry, eddy break-up and laminar flamelet models. The first three combustion models are based on fast chemistry assumption, while the last one is a non-equilibrium model. All the combustion models are assessed using the same turbulence model, numerical schemes, boundary conditions and CFD code, allowing a more direct comparison of the models for turbulence-chemistry interaction. Furthermore, the turbulence model constants are adjusted to achieve a better agreement of the flow field. This strategy is adopted in recognition of the well known fact that the turbulence model is not universal and that fine tuning is necessary to account for differences in flow geometries and situations. It is consistent with the objective of the study, which is to compare the submodels of turbulence-combustion interaction, rather than to test the true predictive accuracy of the turbulence model.
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. In this regard, it is also important to study the influence of radiation heat transfer on the temperature and minor species. With this objective in view, the flamelet based radiation modelling and the flamelet based NO prediction studies are undertaken.

The configurations selected in this study are jet diffusion flames and bluff-body stabilized flames. The reasons behind these selections are that the jet flames have many practical applications and bluff-body burner provides a flame suitable for the study of turbulence-chemistry interactions with a great similarity to practical combustors used in many industrial applications. The bluff-body burner is, therefore, a suitable compromise as a model problem because it has some of the complications associated with practical combustors while preserving relatively simple and well-defined boundary conditions. The selected jet flames also provides a controlled environment for the study of turbulence-chemistry interaction.

1.3 Accomplishments

The following list summarises the important contributions of this work:

- Modification and upgradation of in-house turbulent reacting flow solver research computer codes to adapt with the associated problems of this work.

- Development of flamelet codes and generation of flamelet libraries of different flames for conserved scalar variance and dissipation rate.
• Development of CFD based iterative scheme for the turbulent diffusion flames to account for radiation heat loss from combustion and flame zones.

• Demonstration of the importance of differential diffusion in flamelet modelling of combustion.

• Modelling of transport and chemical kinetics of H₂/N₂ jet flames and comprehensive validation

• Demonstration of the importance of radiation heat loss in flamelet modelling of turbulent combustion.

• Radiation modelling of CO/H₂/N₂ jet flame and comprehensive validation of CFD based flamelet/radiation results for reacting flows.

• Radiation/combustion modelling of H₂/N₂ jet flame and comparison of results

• Radiation modelling of Sydney HM2 flame; analysis of results and comprehensive validation

• Development of NOx modelling strategy and identified the effect of radiation heat loss on NOx formation.

• Identification of the future broad ranges of research possibilities and extended ideas of the present research
CHAPTER 2

Mathematical Models

If a chemically reacting flow is considered, the system at each point in space and time is completely described by specification of pressure, density, temperature, velocity of the flow and concentration of each species. These properties can be changing in time and space. These changes are the result of fluid flow (called convection), chemical reaction, molecular transport (e.g. heat generation, conduction, diffusion and viscosity) and radiation. A mathematical description of flames therefore has to account for each of these processes (Hirschfelder et al. 1964).

Some properties in reacting flows are characterized by the fact that they are conserved. Such properties are the energy, the mass and the momentum. Summation over all the process that change the conserved properties leads to the conservation equations, which describe the changes in reacting flow. Despite the simplifications, a complete set of mathematical formulation should provide the relevant quantities in turbulent flames at different points in space and time with sufficient accuracy. The main focus of this chapter is to present the mathematical tools that describe the interaction between the turbulence and chemistry by employing different thermo-chemical descriptions.

First, the general transport equations of mass, momentum, energy and species concentrations are introduced and the difficulties associated with the solution of the equations for turbulent reacting flows are described. The averaging concept for variable density flows, are discussed and the time-averaged transport equations are presented. Finally, the modelling approaches for the turbulence as well as the combustion models utilised in the present study are described.
2.1 Some Fundamental Definitions

The quantitative treatment of combustion processes requires some understanding of fundamental concepts and definitions:

A chemical reaction is the exchange and/or rearrangement of atoms between colliding molecules. In the course of a chemical reaction, e.g.,

\[ \text{HCN} + \text{OH} \rightarrow \text{CN} + \text{H}_2\text{O}, \]

The atoms (relevant in combustion: C, H, O and N) are conserved; i.e., they are not created or destroyed. On the other hand, molecules (e.g., HCN, OH, CN, and H\(_2\)O) are not conserved. Atoms and molecules are conveniently counted in terms of amount of substance or mole numbers (unit: mole). 1 mole of compound corresponds to \(6.023 \times 10^{23}\) molecules of that compound. Accordingly, the Avogadro’s constant is \(N_A = 6.023 \times 10^{23}\) mol\(^{-1}\). The mole fraction \(x_i\) of species \(i\) denotes the ratio of the mole number \(n_i\) of species \(i\) to the total mole number \(n = \sum n_i\) of the mixture

\[
x_i = \frac{n_i}{n} = \frac{c_i}{c},
\]

Where \(c = n/V\) is the molar density or concentration of the mixture (\(V\) is the system volume) and \(c_i\) is the concentration of species \(i\).

The mass \(m\) is a fundamental property of matter. The mass fraction \(y_i\) of species \(i\) denotes the ratio of the mass \(m_i\) of species \(i\) to the total mass \(m = \sum m_i\) of the mixture

\[
y_i = \frac{m_i}{m} = \frac{\rho_i}{\rho},
\]

The molar mass or molecular weight \(M_i\) (units of e.g., g/mol) of species \(i\) is the mass of 1 mol of species. The mean molar mass of mixture is \(\bar{M}\) (g/mol) denotes an average of molar mass, using the mole fraction as weight

\[
\bar{M} = \sum x_i M_i,
\]

Densities do not depend on the size (extent) of a system. These are intensive properties, and are defined as the ratio of the corresponding extensive properties and the system volume \(V\). Examples are:

Mass density (density) \(\rho = m/V\)
Chapter 2. Mathematical Models

Molar density (concentration) \[ c = \frac{n}{V} \]

It follows easily that the mean molar mass is given by the expression

\[ \frac{\rho}{c} = \frac{m}{n} = \bar{M} \]

2.2 Governing equations in combustion

In reacting fluids we are dealing with various species, mass concentration, molar concentration, mass fraction, mole fraction, and various forms of the equation of state as summarized below:

Species:

\[ \frac{\partial \rho y_i}{\partial t} + \nabla \cdot (\rho \mathbf{u} y_i) = -\nabla \cdot (\mathbf{A} y_i) + \rho \omega_i, \quad i = 1, \ldots, N_s \] (2.1)

Mass:

\[ \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0 \] (2.2)

Momentum:

\[ \frac{\partial \mathbf{u}}{\partial t} + \nabla \cdot (\rho \mathbf{u} \mathbf{u}) = -\nabla p + \nabla \cdot \mathbf{\tau} + \rho \sum_i y_i f_i \] (2.3)

Energy:

\[ \frac{\partial \rho (e + k)}{\partial t} + \nabla \cdot [\rho (e + k) \mathbf{u}] = -\nabla \cdot (\mathbf{\tau} \cdot \mathbf{u}) - \nabla \cdot \mathbf{q} + \rho \sum_i Y_i f_i (u + V_i) \] (2.4)

Viscous Stress:

\[ \mathbf{\tau} = 2\mu \left[ S - \frac{1}{3} (\nabla \cdot \mathbf{u}) I \right] + \mu_B (\nabla \cdot \mathbf{u}) I \] (2.5)

Heat Flux:

\[ q = -2\kappa \nabla T + \sum_i \rho V_i y_i h_i + \dot{\mathbf{k}} T \sum_i \sum_j x_i D_{T,i} (V_i - V_j) + \frac{q_E}{\text{radiation}} \] (2.6)
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Mass Diffusion:

\[ \nabla x_i = \sum_i \frac{x_i x_j}{D_{ij}} (V_j - V_i) + (y_i - x_i) \frac{\nabla p}{p} + \frac{\rho}{p} \sum_j y_i y_j (f_i - f_j) \]

\[ + \sum_j \frac{x_i x_j}{\rho D_{ij}} \left( \frac{D_{T,i}}{y_j} - \frac{D_{T,j}}{y_i} \right) \frac{\nabla T}{T} \]

Stefan-Maxwell pressure-gradient body-force Soret effect

Thermodynamic State:

\[ p = \rho \sum_i \frac{y_i}{M_i} \hat{R} T \]

(2.8)

Where \( V_i \) is the mass diffusion velocity of species \( i \), \( \omega_i \) is the chemical production rate of species \( i \), \( u \) the velocity vector and \( U \) is the normalizing velocity scale, \( y_i \) and \( x_i \) are mass fraction and mole fraction of species \( i \), \( Z \) is the mixture fraction, \( M_i \) is the molecular mass of species \( i \).

Note that summation of all species conservation equations in (2.1) yields total mass conservation, (2.2), so that one of these \( N_s + 1 \) equations is redundant. To be consistent with mass conservation, the diffusion velocities and chemical sources must satisfy

\[ \sum_i y_i V_i = 0, \quad \sum_i \omega_i = 0. \]

(2.9)

Equation (2.7) is an implicit vector equation for the species diffusion velocities. Temperature is implicitly related to internal energy or enthalpy through

\[ e = \sum_i y_i e_i(T), \quad h = e + \frac{p}{\rho} = \sum_i y_i h_i(T) \]

(2.10)

where \( e_i \) and \( h_i \) are the species internal energies and enthalpies per unit mass, which for an ideal gas are functions of temperature only. Species mole and mass fractions are related by

\[ x_i = \frac{y_i/M_i}{\sum_j (y_j/M_j)}, \quad y_i = \frac{M_i x_i}{\sum_j M_j x_j} \]

(2.11)
To complete the specification of the governing equations, molecular transport, thermochemical, and chemical kinetic property data are needed. Transport properties include molecular viscosity $\mu$, thermal conductivity $\kappa$, mass diffusion coefficient $D_{ij}$, and thermal diffusion coefficient $D_{T,i}$ of species $i$, while thermochemical data include $h_i$ and $M_i$. Chemical kinetics will provide the chemical reaction sources, $w_i$, as functions of species concentrations, temperature, and pressure.

The simplified basic set of balance equations comprises the classical Navier-Stokes, species and energy transport equations. These instantaneous local balance equations in classical lettering are:

- **Mass:**
  \[
  \frac{\partial p}{\partial t} + \frac{\partial p u_i}{\partial x_j} = 0 \tag{2.12}
  \]

- **Momentum ($i = 1,2,3$):**
  \[
  \frac{\partial p u_i}{\partial t} + \frac{\partial p u_j u_i}{\partial x_j} = -\frac{\partial p}{\partial x_i} + \frac{\partial \tau_{ij}}{\partial x_j} + F_i \tag{2.13}
  \]
  where $\tau_{ij}$ is the viscous force tensor and $F_i$ a body force.

- **Species ($N$ species with $k = 1,..., N$):**
  \[
  \frac{\partial p Y_k}{\partial t} + \frac{\partial p u_j Y_k}{\partial x_j} = -\frac{\partial J^k_i}{\partial x_i} + \hat{\omega}_k \tag{2.14}
  \]
  $J^k_i$ is the molecular diffusive flux of the species $k$ and $\hat{\omega}_k$ the mass reaction rate of this species per unit volume.

- **Total enthalpy $h_i = h + u_i u_i/2$:**
  \[
  \frac{\partial \rho h_i}{\partial t} + \frac{\partial p u_j h_i}{\partial x_j} = \frac{\partial p}{\partial t} + \frac{\partial J^k_i}{\partial x_i} \left( J^k_i + u_j \tau_{ij} \right) + u_j F_j \tag{2.15}
  \]
  where $u_j \tau_{ij}$ and $u_i F_i$ denote respectively the power due to viscous and body forces. These equations are closed by the expressions for the species molecular fluxes and the
viscous forces. In practical situations, all fluids are assumed to be Newtonian, i.e. the viscous stress tensor $\tau_{ij}$ is given by the Newtonian law:

$$\tau_{ij} = \mu_l \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} - \frac{2}{3} \delta_{ij} \frac{\partial u_k}{\partial x_k} \right)$$  \hspace{1cm} (2.16)$$

where the molecular viscosity $\mu_l$ depending on the fluid properties is introduced. $\delta_{ij}$ is the Kronecker delta symbol.

Species molecular diffusivities are generally described using the Ficks law, assuming a major species:

$$J_i^k = -\frac{\mu_l}{Sc_k} \frac{\partial Y_k}{\partial x_i}$$ \hspace{1cm} (2.17)$$

$Sc_k$ is the Scmidth number of the species $k$, defined as:

$$Sc_k = \frac{\mu_l}{\rho D_k}$$ \hspace{1cm} (2.18)$$

$D_k$ is the molecular diffusivity of the species $k$ relative to the major species.

For multi-species molecular diffusion the expression may be more complex. Soret effect (Species diffusion under temperature gradient) and molecular transport due to pressure gradients are usually neglected. Enthalpy diffusion is described according to Fourier Law:

$$J_i^h = -\frac{\mu_l}{Pr} \left[ \frac{\partial h}{\partial x_i} + \sum_{k=1}^{N} \left( \frac{Pr}{Sc_k} - 1 \right) h_k \frac{\partial Y_k}{\partial x_i} \right]$$ \hspace{1cm} (2.19)$$

The Prandtl number $Pr$ compares the diffusive transport of momentum (viscous force) and temperature. In the previous expression, the radiative heat transfer and Dufour effect (energy transport caused by density gradients) are neglected. The Prandtl number is written as a function of the thermal diffusivity $\lambda$ and the constant pressure specific heat $C_p$:
\[ Pr = \left( \frac{\mu C_p}{\lambda} \right) \quad (2.20) \]

Then, the Lewis number \( L_e_k \) of the species \( k \), comparing thermal and mass diffusivities is introduced:

\[ L_e_k = \left( \frac{Sc_k}{Pr} \right) = \left( \frac{\lambda}{\rho C_p D_k} \right) \quad (2.21) \]

Under the assumption of unity Lewis number, the enthalpy diffusive flux (Eq. 2.19) is simplified and mass fraction and enthalpy balance equations are formally identical if \( \partial p/\partial t \), \( u_i \tau_{ij} \) and \( u_i F_i \) are negligible (low Mach number assumption) Poinso\-t and Veynante (2001). The term \( \partial p/\partial t \) is negligible in many practically occurring situations, e.g. jets, gas turbines and furnaces (Jones and Khaki 1996). This assumption is generally made to simplify turbulent flame modelling, especially in premixed flames when species mass fractions and temperature are assumed to be equivalent variables. Nevertheless, thermo-diffusive instabilities occur in premixed systems when the Lewis number is lower than unity (for example for hydrogen). One direct consequence of these instabilities is an increase of the premixed flame area and of the global reaction rate as described by Williams (1985) and Trouvé and T. Poinso\-t (1994).

In combusting flows many species with widely disparate molecular weights participate with the consequence that the proper treatment of the molecular fluxes involves the full array of diffusion coefficients characterising the diffusion of species \( \alpha \) into a mixture involving species \( i \), \( (i=1,2,\ldots,N) \) (Libby and Williams 1994). However, when simplified form of energy equation is applied for turbulent flows, the complete treatment of the molecular diffusion flux is less important and Fick’s law of diffusion \( J_i^k = -\frac{\mu_i}{Sc_k} \frac{\partial Y_i}{\partial x} \) is used to represent the molecular diffusion flux.

To obtain the chemical source term, a system involving \( N \) chemical species and \( R \) reaction steps is considered, of the form (Libby and Williams 1994):
where $M'_a$ is the chemical symbol of species $\alpha$, $\nu'_{a\beta}$ and $\nu''_{a\beta}$ are the stoichiometric coefficients of species $\alpha$ in the reaction step $\beta$. The chemical source term $\dot{\omega}_\alpha$ is the net result of the construction and destruction of species $\alpha$ due to each reaction step $\beta$, i.e.

$$\dot{\omega}_\alpha = \sum_{\beta=1}^{R} \dot{\omega}_{a\beta}$$

where $\dot{\omega}_{a\beta} = 0$ if species $\alpha$ does not participate in the reaction step $\beta$. In any particular reaction step, the rate of formation of species $\alpha$, is the net rate of formation of $\alpha$ due to forward and backward reaction steps in reaction $\beta$:

$$\dot{\omega}_{a\beta} = M_{a\beta} \left( \nu''_{a\beta} - \nu'_{a\beta} \right) \times$$

$$\left\{ k_{f\beta} \rho^{m_{\beta}} \prod_{a=1}^{N} \left( \frac{Y_{\alpha}}{M_{\alpha}} \right)^{\nu''_{a\beta}} - k_{b\beta} \rho^{l_{\beta}} \prod_{a=1}^{N} \left( \frac{Y_{\alpha}}{M_{\alpha}} \right)^{\nu'_{a\beta}} \right\}$$

where $m_{\beta} = \sum_{i=1}^{N} \nu'_{a\beta}$ and $l_{\beta} = \sum_{i=1}^{N} \nu''_{a\beta}$ are the orders of the forward and backward reactions respectively. $k_{f\beta}$ and $k_{b\beta}$ are the Arrhenius rate coefficients of the forward and backward reactions. The rate coefficients are expressed as:

$$k_{f\beta} = B_{\beta} T^{a_{\beta}} \exp \left( -\frac{E_{\beta}}{\hat{R}T} \right)$$

where $B_{\beta}$ is the pre-exponential factor, $a_{\beta}$ is a constant exponent, $E_{\beta}$ is the activation energy and $\hat{R}$ is the universal gas constant.

Combustion influences the flow field through density and the equation of state for ideal gases is used to express density:

$$P = \rho \hat{R} T \sum_{\alpha=1}^{N} \frac{Y_{\alpha}}{M_{\alpha}}$$
The equations described so far constitute a closed set of equations and in principle they can be solved numerically with appropriate initial and boundary conditions. However, the direct numerical simulation (DNS) requires large computer memory and CPU time to resolve very small spatial and time scales. For example, the number of grid points needed in the DNS is proportional to $Re_t^{9/4}$, where $Re_t$ is the turbulence Reynolds number, for three dimensional inert mixing layer flows (Libby and Williams 1994). The LES (large eddy simulation) may be the better alternative but its application to combustion modeling is still at an early stage. As in RANS, the complex coupling between micromixing and chemical reactions occurring at unresolved scales needs models, however, LES possesses some attractive properties.

### 2.3 Reynolds and Favre averaging

The full numerical solution of the instantaneous balance equations is limited to very simplified cases (Poinsot (1996), Vervisch and Poinsot (1998)), where the number of time and length scales present in the flow is not too great. To overcome this difficulty, an additional step is introduced by averaging the balance equations to describe only the mean flow field (local fluctuations and turbulent structures are integrated in mean quantities and these structures have no longer to be described in the simulation). Each quantity $Q$ is split into a mean $\bar{Q}$ and a deviation from the mean denoted by $Q'$:

$$Q = \bar{Q} + Q' \quad \text{with} \quad \bar{Q}' = 0 \quad (2.27)$$

Then, the governing equations may be ensemble averaged to derive transport equations for the mean quantity $\bar{Q}$. This classical Reynolds averaging technique, widely used to study non-reacting fluid flows, brings unclosed correlations like $u'Q'$ which are unknown and must be modeled. This numerical procedure is called Reynolds Averaged Navier–Stokes (RANS) modeling.

In the study of turbulent flames, fluctuations of density are observed because of the thermal heat release, and there are some additional difficulties in Reynolds averaging. Simply, averaging the mass balance equation leads to:

$$\frac{\partial \bar{\rho}}{\partial t} + \frac{\partial}{\partial x_i} \left( \rho \bar{u}_i + \rho' u_i' \right) = 0 \quad (2.28)$$
where the velocity/density fluctuations correlation $\overline{\rho' u'_i}$ appears. To avoid the explicit modeling of such correlations, a Favre (mass weighted) average is introduced according to Favre (1969) and any quantity is then decomposed into $\overline{Q} = \bar{Q} + Q^*$:

$$\bar{Q} = \frac{\rho \overline{Q}}{\rho}; \quad \text{and} \quad Q^* = \frac{\rho (Q - \bar{Q})}{\rho} = 0 \quad (2.29)$$

The Favre averaged continuity equation:

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho \bar{u}_i}{\partial x_i} = 0 \quad (2.30)$$

Is then formally identical to the Reynolds averaged continuity equation for constant density flows. This result is true for any balance equations (momentum, energy, mass fractions, etc.). Nevertheless, Favre averaging is only a mathematical formalism:

- There is no simple relation between Favre, $\bar{Q}$ and Reynolds, $\overline{Q}$, averages. A relation between $\bar{Q}$ and $\overline{Q}$ requires the knowledge of density fluctuations correlations $\overline{\rho' Q'}$ remaining hidden in Favre averaging:

$$\bar{\rho} \bar{Q} = \rho \overline{Q} + \overline{\rho' Q'} \quad (2.31)$$

- Comparison between numerical simulations, providing Favre averaged quantities $\bar{Q}$, with experimental results are not obvious. Most experimental techniques determine Reynolds averaged data $\overline{Q}$ and difference between $\bar{Q}$ and $\overline{Q}$ may be significant.

### 2.4 Favre averaged balance equations

Applying Favre averaging to instantaneous balance equations yields:
• Mass:

\[ \frac{\partial \rho}{\partial t} + \frac{\partial \rho \vec{u}_j}{\partial x_j} = 0 \]  

(2.32)

• Momentum \((i = 1, 2, 3):\)

\[ \frac{\partial}{\partial t} (\bar{\rho} \vec{u}_i) + \frac{\partial}{\partial x_j} (\bar{\rho} \vec{u}_j \vec{u}_i) = -\frac{\partial \bar{p}}{\partial x_i} - \frac{\partial}{\partial x_j} (\bar{\rho} \vec{u}_j \vec{u}_i^* \vec{u}_j^*) + \frac{\partial \tau_{ij}}{\partial x_j} + \bar{F}_i \]  

(2.33)

• Chemical species (for \(N\) species, \(k = 1, \ldots, N\)):

\[ \frac{\partial}{\partial t} (\bar{\rho} \bar{Y}_k) + \frac{\partial}{\partial x_j} (\bar{\rho} \bar{u}_j \bar{Y}_k) = -\frac{\partial}{\partial x_j} (\bar{\rho} \vec{u}_i \vec{u}_j Y_k^*) + \bar{\omega}_k \]  

(2.34)

• Total enthalpy, \(\bar{h}\):

\[ \frac{\partial}{\partial t} (\bar{\rho} \bar{h}) + \frac{\partial}{\partial x_j} (\bar{\rho} \bar{u}_j \bar{h}) = -\frac{\partial}{\partial x_j} (\bar{\rho} \vec{u}_i \vec{u}_j h_i^*) + \bar{\mu}_j \frac{\partial \bar{h}}{\partial x_j} + \bar{\tau}_{ij} \left( J_i^k + \bar{u}_i \bar{\tau}_{ij} \right) + \bar{F}_{ij} \]  

(2.35)

The objective of the turbulent combustion modeling is to propose closures for the unknown quantities appearing in the averaged balanced equations, such as:

• Reynolds stresses \(\vec{u}_i \vec{u}_j^*\). The turbulent model provides an approximation for this term. The closure may be done directly or by deriving balance equations for these Reynolds stresses. However, most combustion works are based on turbulent modelling developed for non-reacting flows, such as \(k - \varepsilon\), simply written in terms of Favre averaging, and heat release effects on the Reynolds stresses are generally not explicitly included.

• Species \((\bar{u}_i \bar{Y}_k^*)\) and temperature \((\bar{u}_i \bar{T})\) turbulent fluxes. These fluxes are usually closed using a gradient transport hypothesis:

\[ \bar{\rho} \vec{u}_i \bar{Y}_k^* = \frac{\mu_t}{S_{C_{\kappa}}} \frac{\partial \bar{Y}_k}{\partial x_j} \]  

(2.36)

where \(\mu_t\) is the turbulent viscosity, estimated from the turbulence model, and \(S_{C_{\kappa}}\) a turbulent Schmidt number for the species \(k\).
• Laminar diffusive fluxes $J^i_t, J^h_t$, etc. are usually small compared to turbulent transport, assuming a sufficiently large turbulence level (large Reynolds numbers limit).

• Species chemical reaction rates $\bar{\omega}_k$. Turbulent combustion modelling generally focuses on the closure of these mean burning rates.

These equations, closed with appropriate models, allow only for the determination of mean quantities that may differ from instantaneous ones. Strong unsteady mixing effects, resulting from the rolling up of shear layers, are observed in turbulent flames, and the knowledge of steady statistical means is not always sufficient to describe turbulent combustion.

2.5 Turbulence model

Several models have been proposed to model Reynolds stress. Among these, the relatively simple $k-\varepsilon$ model has been widely used for turbulent combustion calculations (Brookes and Moss 1999; Biagioli 1997; Sandlers and Lamers 1994; Liew et al. 1984; Lentini 1994). The $k-\varepsilon$ model has shown a number of shortcomings, such as its inability to correctly represent the anisotropy of the Reynolds stresses and its overprediction of the spreading rate of jet and bluff body flows (Dally et al. 1998b). Many of these shortcomings are eliminated in higher order closure formulations such as second moment closure and Reynolds-stress closure. Over the last few years, several second moment closure models have been developed and applied in a number of turbulent combusting flows (Jones 1994). However, the simple $k-\varepsilon$ model can capture the characteristics of flow fields of bluff body flames reasonably accurately (Dally et al. 1998b). The present study focus on the prediction of the thermo-chemistry in jet and bluff body flames by different chemistry models. For that purpose, the $k-\varepsilon$ description is sufficient.

Most of the work in turbulence modelling is based on constant density flows (Jones and Whitelaw 1982). The modelling of inert turbulent flows is significantly complex without the inclusion of the variable density contributions introduced by combustion. In turbulent combustion, large density fluctuations arise and must be accounted for in the modelling. The common approach for modelling variable density flows is to recast the
Reynolds averaged model in terms of Favre-averaged quantities with the assumption that the density variations are accounted for by averaging. The extension of the constant density $k$-$\varepsilon$ model for variable density flows is described in Kuo (1986) and Jones and Whitelaw (1982). The starting point for modelling the Reynolds stress is the eddy viscosity model:

$$
\rho \bar{u}_i \bar{u}_j = -\mu_t \left( \frac{\partial \bar{u}_i}{\partial x_j} + \frac{\partial \bar{u}_j}{\partial x_i} \right) + \frac{2}{3} \rho \delta_{ij} \frac{\partial k}{\partial x_i} + \frac{2}{3} \mu_t \frac{\partial \bar{u}_i}{\partial x_i} \delta_{ij}
$$

(2.37)

The turbulent viscosity $\mu_t$ is assumed to be proportional to the product of a turbulent velocity scale and a length scale. In the $k$-$\varepsilon$ model, these velocity and length scales are obtained from two parameters, the turbulent kinetic energy, $k$, and the dissipation rate of turbulent kinetic energy, $\varepsilon$. The velocity scale is taken to be $\sqrt{k}$ and the length scale is taken to be $\sqrt{\frac{k^3}{\varepsilon}}$. Thus, the eddy viscosity $\mu_t$ is given by:

$$
\mu_t = \bar{\rho} C_\mu \frac{k^2}{\varepsilon}
$$

(2.38)

where $C_\mu$ is a proportionality constant. The modelled equations for the turbulent kinetic energy and energy dissipation rate are given by:

$$
\frac{\partial}{\partial x_j} (\bar{\rho} \bar{u}_i \bar{k}) = \frac{\partial}{\partial x_j} \left( \frac{\mu_t}{\sigma_k} \frac{\partial \bar{k}}{\partial x_j} \right) + G - \bar{\rho} \bar{\varepsilon}
$$

(2.39)

and

$$
\frac{\partial}{\partial x_j} (\bar{\rho} \bar{u}_i \bar{\varepsilon}) = \frac{\partial}{\partial x_j} \left( \frac{\mu_t}{\sigma_k} \frac{\partial \bar{\varepsilon}}{\partial x_j} \right) + C_{\varepsilon_1} \frac{\bar{\varepsilon}}{k} G - C_{\varepsilon_2} \bar{\rho} \frac{\bar{\varepsilon}^2}{k}
$$

(2.40)

where $C_{\varepsilon_1}$, $C_{\varepsilon_2}$ are the empirical constants and $\sigma_k$ and $\sigma_\varepsilon$ are the Prandtl numbers governing the turbulent diffusion of $k$ and $\varepsilon$, $G$ is the rate of production of turbulent kinetic energy:

$$
G = \mu_t \left( \frac{\partial \bar{u}_j}{\partial x_i} + \frac{\partial \bar{u}_i}{\partial x_j} \right) \frac{\partial \bar{u}_i}{\partial x_i}
$$

(2.41)
The constants are calibrated in simple (constant density) flows, such as decaying grid turbulence ($\Rightarrow C_{c2}$), near wall flows (assuming production=dissipation and $\overline{u'v'}/k \approx \text{constant} \Rightarrow C_{c1}$, $C_{\mu}$). The constants are assigned values with computer optimisation so that the widest range of flows possible can be predicted. The following values are recommended: $C_{\mu} = 0.9$; $C_{c1} = 1.44$; $C_{c2} = 1.92$; $\sigma_{k} = 1.0$; $\sigma_{e} = 1.3$. For free flows $\sigma_{f} = 0.7$ and near wall flows $\sigma_{f} = 0.9$ (Libby 1996).

The standard $k$-$\varepsilon$ model does not produce the correct spreading rate of a single axisymmetric jet issuing into a co-flowing or stagnant environment. Several empirical modifications of the model constants have been proposed to remedy this shortcoming (McGuirk and Rodi 1979; Pope 1978; Thies and Tam 1996). The overprediction of the spreading rate is also observed for the case of bluff body flames (Gran and Magnussen 1996a; Correa and Gulati 1992; Gran et al. 1997). The modifications similar to those for the jet flow and also a simple modification in the value of $C_{c1}$, i.e. $C_{c1} = 1.60$ have been implemented for a bluff body flame (Dally et al. 1998b). They have shown that the simple modification of $C_{c1}$ is sufficient to produce a good prediction of the spreading rate of the jet and the bluff body flows. The value of $C_{c1}$ has been adjusted from 1.44 to 1.60 and has been implemented as standard in the third TNF workshop (TNF3 1998).

### 2.6 Combustion models

Averaging of the species concentration equation leads to the mean reaction rate term (see Eq.2.34). Direct closure of the mean reaction rate term through Taylor series expansion is not feasible for highly exothermic combustion reactions (Williams 1985). No suitable deterministic method is available to date to determine the mean reaction rate term. However, a number of combustion models are available, which do not require the specification of the mean reaction rate. These models reduce the turbulent reacting flow problem to a tractable form by introducing a single scalar that characterises the thermo-chemistry of the reacting mixture.
2.6.1 Conserved scalar models

In conserved scalar models of non-premixed combustion, the instantaneous thermo-chemical state of the mixture is determined by a single conserved scalar. Detailed formulation and the analysis of the conserved scalar model can be found in many texts, e.g. Kuo (1986), Williams (1985) and more recently in Warnatz et al. (1996). With the assumptions of equal diffusivity, unity Lewis number and adiabatic combustion, all the conserved scalars (elemental mass fractions and enthalpy) can be related to a single normalised conserved scalar, the mixture fraction. Thermo-chemical models are then required to specify the relationship between the mixture fraction and the scalar variables (temperature, species concentrations). These models are based on the "fast" chemical reaction, e.g. the time required to complete reaction is much shorter than the convection and diffusion time in the turbulent flame. A brief description of the conserved scalar models is given in following sections.

Flame sheet model

This is the simplest reaction scheme which assumes that the reaction takes place in a thin flame sheet at an infinitely fast rate. The reaction scheme is called "mixed-unsburned" approximation. The flame sheet is located at the stoichiometric mixture fraction. The chemistry is infinitely fast and irreversible, with fuel and oxidant species never coexisting in space and complete one-step conversion to final products. This description allows species concentrations to be determined directly from the given reaction stoichiometry, with no reaction rate or chemical equilibrium information required. This simple system description yields straight line relationships between the species mass fractions and the mixture fraction, as shown in Figure 2.1.
The model calculates the species concentrations and temperature from algebraic equations based on the reaction stoichiometry, without any information of reaction rate or equilibrium condition. The species concentrations and temperature are calculated from (Lockwood and Naguib 1975; Chung 1993):

\[
0 \leq Z \leq Z_{st} \quad Y_{fu} = 0 \quad Y_{ox} = Y_{ox,A} \frac{Z_{st} - Z}{Z_{st}} \quad (2.42)
\]

\[
Z_{st} \leq Z \leq 1.0 \quad Y_{ox} = 0 \quad Y_{fu} = Y_{fu,F} \frac{Z - Z_{st}}{1 - Z_{st}} \quad (2.43)
\]

where, the mixture fraction is defined as:

\[
Z = \frac{(sY_{fu} - Y_{ox}) + Y_{ox,A}}{sY_{fu} + Y_{ox,A}} \quad (2.44)
\]

and the stoichiometric mixture fraction as:
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\[ Z_{st} = \frac{Y_{ox,A}}{sY_{fu,F} + Y_{ox,A}} \]  

(2.45)

Here, \( Y_{fu} \) and \( Y_{ox} \) denote the mass fraction of fuel and oxidiser respectively; \( s \) is the stoichiometric oxygen required burning 1 kg of fuel and the subscripts A and F denote the air and fuel stream respectively.

The mass fraction of the product can be obtained from the conservation of mass as:

\[ Y_{pr} = 1.0 - (Y_{ox} + Y_{fu}) \]  

(2.46)

For adiabatic combustion, the enthalpy is also a conserved scalar and with the assumption of unity Lewis number, the instantaneous enthalpy and thermo-chemical properties are related to the instantaneous mixture fraction according to the relations:

\[ h(Z) = Z h_F + (1 - Z) h_A \]  

(2.47)

\[ \int_0^Z \bar{C}_p dT = h(Z) - Y_{fu} H_{fu} \]  

(2.48)

\[ C_p (Z) = \sum_{\alpha} Y_{\alpha} (Z) C_{p\alpha} (Z) \]  

(2.49)

\[ \rho(Z) = \frac{P W(Z)}{R T(Z)} \]  

(2.50)

\[ \frac{1}{W(Z)} = \frac{Y_{fu}(Z)}{W_{fu}} + \frac{Y_{ox}(Z)}{W_{ox}} + \frac{Y_{pr}(Z)}{W_{pr}} \]  

(2.51)

where, \( H_{fu} \) is the heating value of fuel, \( C_p \) is the specific heat and \( W \) is the molecular weight and \( R \) is the universal gas constant. An obvious shortcoming of the flame sheet model is that it does not calculate intermediates, minor species and radicals. In many applications, it is desirable to have an insight into the behaviour of these species. Moreover, the absence of these radicals in the mixture can give a substantial
underestimation of the specific heat, which consequently leads to an overprediction of the temperature. The flame sheet model was developed in the early 70's. Some of the earlier work can be found in Elghobashi and Pun (1974), Lockwood et al. (1974) and Elghobashi (1979). Because of its simplicity, the flame sheet model is still being used in combustion calculations (Dally et al. 1998a; Chung 1993). In applications where only the mean flame structure, temperature and major species are required without precise accuracy, this model is a good choice as the model is very simple and subsequently, required less computational resources.

Equilibrium/Constrained equilibrium models
The gas mixture is assumed to be at chemical equilibrium at every location and time in the flame for equilibrium model. The equilibrium composition and temperature of the flame are calculated as a function of mixture fraction based on the minimization of Gibbs free energy (Kuo 1986; Warnatz et al. 1996). An important advantage of the chemical equilibrium model is that it can provide information about the minor species and radicals without a detailed knowledge of the reaction kinetics.

The chemical equilibrium model is valid only if the characteristic Damköhler numbers are much larger than unity, e.g. the reaction processes are much faster than the convection and diffusion processes of the flow. However, in many practical and laboratory flames, only part of the reaction processes are fast enough to reach chemical equilibrium. The slower, lower Damköhler number reactions interact with the convection and diffusion processes of the flows. Such finite rate chemistry effects lead to the abundance of the species involved. The mass fraction of these species can not be adequately represented by the equilibrium model. A well known example of this is provided by the super-equilibrium concentrations of OH radicals found in the upstream part of turbulent flames (Drake et al. 1984; Masri et al. 1996; Barlow et al. 1990). Here, the consumption of OH is governed by three body recombination reactions, which are relatively slow and do not reach the equilibrium condition in the upstream part of flames where intense mixing occurs. The resultant balance of production, consumption, convection and diffusion is such that the OH level can be in excess of the equilibrium level by a factor of 5.
The chemical equilibrium approach has been widely used and reasonable temperature and major species concentrations can be obtained. The major shortcoming of the equilibrium model is that related to its prediction of unrealistically high level of CO in the fuel rich mixture of hydrocarbon flames (Jones 1980). The constrained equilibrium model is an empirical modification to the equilibrium model to reproduce the realistic level of CO in the fuel rich mixture. The model is based on the idea that reactions in hydrocarbon flames occur primarily in a small zone around the stoichiometric mixture fraction. Outside this zone, if the temperature is too low or the mixture is too rich or lean, the fuel will not burn. In the calculation, the equilibrium condition is applied for mixture leaner than the rich flammability limit. In case of mixture richer than the rich flammability limit, the flame is extinguished and the unburned fuel mixes with the equilibrium composition at the rich flammability limit.

An alternative constrained equilibrium model is also available (Bilger and Starner 1983) where the thermochemistry is described by a reaction zone at the stoichiometric mixture fraction and by a fuel breakup/pyrolysis sheet at a slightly higher mixture fraction. A portion of the hydrocarbon fuel breaks up into intermediate hydrocarbons in a one-step, irreversible and infinitely fast reaction at the fuel breakup/pyrolysis sheet. These intermediates are considered to be consumed at the stoichiometric mixture fraction surface. The concentration of other species are computed from the equilibrium approximation taking into account the constraints for the fuel and the intermediate species. The constraints imposed on the reacting system are assumed to be a linear combination of species compositions present in a given system. They can be written in the form

\[ C_i = \sum_{j=1}^{n_s} a_{ij} N_j \quad i = 1, \cdots, n_c \] (2.52)

where \( a_{ij} \) is the value of the constraint \( i \) for the species \( j \) and \( n_c \) is the number of constraints. The rate equations for the constraints can easily be determined by differentiating the above equation (2.52):

\[ \dot{C}_i = \sum_{j=1}^{n_s} a_{ij} \dot{N}_j \quad i = 1, \cdots, n_c \] (2.53)
And using the species rate equations

\[ \dot{N}_j = V \sum_{k=1}^{n_r} v_{jk} r_k \quad j = 1, \cdots, n_s \]

\[ \dot{C}_i = V \sum_{k=1}^{n_r} b_{ik} r_k \quad i = 1, \cdots, n_c \]  \hspace{1cm} (2.54)

where the \( N_j \) is the number of moles of species \( j \), \( V \) is the volume of the system, \( v_{jk} = v_{jk}^+ - v_{jk}^- \) is the net change in moles of species \( j \) due to reaction \( k \), \( r_k = r_k^+ - r_k^- \) is the net reaction rate per unit volume, and \( r_k^+ \) and \( r_k^- \) are the forward and reverse reaction rates.

where \( b_{ik} = V \sum_{j=1}^{n_s} a_{ij} v_{jk} \) is the change of constraint \( i \) due to the reaction \( k \) and \( n_b \) is the number of reactions which change the constraints, that is for which \( b_{ik} \neq 0 \).

Since elements are conserved their corresponding constraints are constant, then for elements, \( b_{ik} = 0 \) and \( \dot{C}_i = 0 \) for \( i = 1, \cdots, n_e \) where \( n_e \) is the number of elements in the system. This ensures conservation of elements and reduces the number of rate equations to be integrated by \( n_e \).

Given the initial conditions the \( n_c - n_e \) rate equations (2.53) for the constraints can be integrated in stepwise fashion using standard integration routines. At each step the constrained-equilibrium composition

\[ N_j(t) = N_j(T(t), V(t), C_1(t), \cdots, C_{n_e}(t), \cdots, C_{n_c}(t)) \]  \hspace{1cm} (2.55)

must be evaluated as a part of the constrained-equilibrium computation.

The constrained-equilibrium composition is found by maximizing the entropy or minimizing the Gibbs free energy of the mixture subject to a set of constraints using the Lagrange multipliers method. For each constraint there is a corresponding Lagrange multiplier (constraint potential). The species composition is then determined as a function of those multipliers:
\[ N_j = Q_j \exp \left( -\sum_{i=1}^{n_c} a_y \gamma_i \right) \quad j = 1, \ldots, n_s \]  

(2.56)

where \( Q_j \frac{P_0 V}{RT} \exp(-\mu_j^o) \) is the partition function for species \( j \), and \( \mu_j^o = \frac{F_j^o}{RT} \) is the dimensionless standard Gibbs free energy and \( \gamma_i \) is the Lagrange multiplier (constraint potential) conjugate to the \( i \)th constraint. The system is described by \( n_c \) constraint potentials instead of \( n_s \) species, thus the order of the system is reduced by \( n_s - n_c \), which results in a reduction of the number of rate equations to be integrated. Once the constraint potentials \( \gamma_i \) have been determined the constrained composition can easily be calculated using equation (2.56).

In turbulent diffusion combustion the fast chemistry assumption approach which can be used conveniently in some combustion modelling cannot be extended to situations where the finite rate chemistry are known to have appreciable effects. For example, oxidation of CO in gas turbine engines, formation of trace species (NO and soot) and extinction conditions. In such cases the description involves a second variable to be integrated, namely the progress variable. The closure of turbulence chemistry interaction involves solution for the transport equations of the mean value of progress variable together with its fluctuations from which the joint PDF of the mixture fraction and progress variable can be determined. The temperature and thermo-chemical variables are averaged using this PDF and made available to the calculations through a look-up table, which is a systematic arrangement of a scalar variable or an instantaneous property data in the flow domain as function of mean mixture fraction \( \bar{f} \) and mixture fraction variance \( \bar{f}^{r2} \). For adiabatic single mixture fraction lookup table the scaled mixture fraction variance \( \bar{f}_{s}^{r2} = 0.25 \frac{\bar{f}^{r2} f_{\text{fuel}}^{r2}}{f f (1-f)} \) is used instead of \( \bar{f}^{r2} \) as discussed in Fluent manual (1999). For a system of two mixture fractions there will be lookup table for each instantaneous scalar property \( \phi_i \) as a function of fuel mixture fraction \( f_{\text{fuel}} \) and the secondary partial fraction \( p_{\text{sec}} \) that is.
\[ \phi_i = \phi_i(f_{\text{fuel}}, P_{\text{sec}}) \]  
\[ (2.57) \]

\[ \phi_i = \phi_i(f_{\text{fuel}}, P_{\text{sec}}, H^*) \]  
\[ (2.58) \]

The lookup table formats for adiabatic and nonadiabatic conditions have been shown in table 2.1

<table>
<thead>
<tr>
<th>Type of model</th>
<th>Adiabatic</th>
<th>Non adiabatic</th>
</tr>
</thead>
<tbody>
<tr>
<td>Single mixture fraction</td>
<td>( \overline{f}, \overline{f}_M )</td>
<td>( \overline{f}, \overline{f}_M, \overline{H^*} )</td>
</tr>
<tr>
<td>Two mixture fractions</td>
<td>( f_{\text{fuel}}, f_{\text{sec}} )</td>
<td>( f_{\text{fuel}}, f_{\text{sec}}, H^* )</td>
</tr>
</tbody>
</table>

Where \( H^* \) is the instantaneous enthalpy.

The presumed shape pdf method

The simplest approach to the mixing between fuel and oxidiser in a nonpremixed flame is to presume the shape of the pdf. The nonlinearity of the instantaneous relationship between the mixture fraction and the scalar variables, \( \varphi(Z) \), implies that the mean scalar variables in a turbulent field cannot be obtained from \( \bar{\varphi} = \varphi(\bar{Z}) \). The mean scalar variables in a turbulent field are thus obtained by introducing a probability density function \( P(Z) \) (Jones and Whitelaw 1982):

\[ \bar{\varphi} = \int \varphi(Z)P(Z)dZ \]  
\[ (2.59) \]

The shape of the probability density function is presumed and is given in terms of the mean, \( \bar{Z} \), and variance, \( Z'^2 \), of the mixture fraction. The values of the mean and variance of mixture fraction are obtained from their respective modelled transport equations:

\[ \frac{\partial}{\partial x_j} \left( \bar{\rho} \bar{u}_i \bar{Z} \right) = \frac{\partial}{\partial x_j} \left( \frac{\mu_l}{\sigma_k} \frac{\partial \bar{Z}}{\partial x_j} \right) \]  
\[ (2.60) \]
\[
\frac{\partial}{\partial x_j} (\rho \tilde{u}_j \tilde{Z}^{n_2}) = \frac{\partial}{\partial x_j} \left( \frac{\mu_t}{\sigma_k} \frac{\partial \tilde{Z}^{n_2}}{\partial x_j} \right) + C_{g1} \mu_t \left( \frac{\partial \tilde{Z}^{n_2}}{\partial x_j} \right)^2 - C_{g2} \frac{\tilde{\varepsilon}}{k} \rho \tilde{Z}^{n_2} \tag{2.61}
\]

where \( C_{g1} \) and \( C_{g2} \) are model constants with values of 2.0 and 2.8 respectively. The common types of assumed pdf shape used in combustion calculations are the double delta pdf (Khalil et al. 1975), clipped Gaussian distribution (Lockwood and Naguib 1975) and beta pdf (Jones and Priddin 1978). In a comparative study of the above pdf functions, Jones (1980) has shown that the double delta function gives unrealistic double peak values in the radial temperature profiles. Both the clipped Gaussian and beta pdfs have produced similar results and accurately predicted temperature and species mass fraction profiles. The implementation of the clipped Gaussian function requires an iterative procedure. The beta pdf does not require arbitrary clipping in order to satisfy the bounds of the mixture fraction as with the clipped Gaussian pdf and the implementation is particularly straightforward. Hence, in the present study the pdf is presumed to have the shape of a beta-function.

### 2.6.2 Laminar flamelet model

In combustion models where nonequilibrium effects are important, the assumption of local chemical equilibrium can lead to unrealistic results. At the molecular level, fuel and oxidizer diffuse into the reaction zone where they encounter high temperatures and radical species and ignite. More heat and radicals are generated in the reaction zone, and some diffuse out. In near-equilibrium flames, the reaction rate is much faster than the diffusion rate. However, as the flame is stretched and strained by the turbulence, species and temperature gradients increase, and radicals and heat more quickly diffuse out of the flame. The species have less time to reach chemical equilibrium, and the local non-equilibrium increases. The laminar flamelet model is suited to predict moderate chemical nonequilibrium in turbulent flames due to aerodynamic straining by the turbulence. The chemistry, however, is assumed to respond rapidly to this strain, so as the strain relaxes, the chemistry relaxes to equilibrium. Typical cases in which the equilibrium assumption breaks down are modeling the rich side of hydrocarbon flames, predicting the intermediate species that govern NOx formation, and modeling lift-off
and blow-off phenomena in jet flames. Several approaches are available to overcome these modeling difficulties on a case-by-case basis; Flamelet models have been proposed as a more general solution to the problem of detailed nonequilibrium flame chemistry.

The laminar flamelet model views the turbulent flame as consisting of an ensemble of stretched laminar flamelets is due to Williams (1975). Later, Liew et al. (1981) proposed using profiles taken from laminar diffusion flames to calculate means and variances in turbulent flames. Flamelet equations based on the mixture fraction as independent variable, using the scalar dissipation rate for the mixing process, were derived and a first review of diffusion flamelet models was given by Peters (1984). For premixed and diffusion flames the flamelet concept was reviewed by Peters (1986) and Bray and Peters (1994)

Flamelets are thin, reactive-diffusive layers embedded within an otherwise nonreacting turbulent flow fields (Figure 2.2). The laminar flamelet model has been presented as a non-equilibrium version of the classical Burke-Schumann limit (Bray and Peters 1994). The approach is based on the notion that, if the chemical time scales are much shorter than the characteristic turbulence time scales, reaction takes place in locally thin one dimensional structures. These structures are assumed to be the same as available in a laminar flame. The main advantage of the flamelet concept lies in the fact that nonequilibrium effects can be incorporated, while the detailed chemistry need not be resolved together with the turbulent flame calculation. The flamelet structure can be conveniently presented in mixture fraction space. Once the flamelet structure has been resolved as a function of the prescribed parameters, and is available in the form of a flamelet library, all scalars (i.e., temperature, density, and species concentrations) are known as functions of the parameters.

An example of a flame that can be entirely mapped into mixture fraction space is a counterflow diffusion flame. Counterflow diffusion flames can be studied quite easily (both numerically and by experiment). They have already been used by many authors to construct libraries of flamelets. The balance equations, solution methods, and sample calculations of the counterflow laminar diffusion flame can be found in several references, and a comprehensive review and analysis is presented in Dixon (1990). The strain rate appears as a parameter in the laminar flamelet equations, and depending on
the imposed boundary conditions, will be either prescribed or calculated as an eigenvalue of the problem Bray and Peters (1994).

In non-premixed turbulent combustion, the reaction zone is attached to the high temperature region close to stoichiometric mixture and advected and diffused with the mixture fraction field (Bray and Peters 1994). Consequently, the flame fronts encountered in the non-premixed flames have no intrinsic, flow field dependent length scale; the flame thickness is determined solely by the mixture fraction field. Based on these observations, Bray and Peters (1994) and Peters (1991) have introduced a phase diagram for the regimes of non-premixed turbulent combustion using two parameters. The first parameter is the Damköhler number and the second one is the mixture fraction
fluctuations around the flame front. The separated flamelets exist when the turbulent fluctuations of mixture fraction are higher than the flame thickness in the mixture fraction space; otherwise connected reaction zones exist. The flamelet regime is observed when the value of the Damköhlar number is greater than one and the mixture fraction variance along the flame front is greater than the flame thickness in the mixture fraction space.

As stated earlier, the laminar flamelet model is presented as a non-equilibrium model. The scalar dissipation rate is identified as a characteristic quantity to describe the departure from chemical equilibrium, Bray and Peters (1994). Based on an asymptotic analysis of a flamelet structure for quasi-steady unity Lewis number flames, Bray and Peters (1994) have concluded that the scalar dissipation rate should account for non-equilibrium effects caused by both convection and diffusion, Bray and Peters (1994). Some other effects, such as rapid unsteady changes, large local differences between convection and diffusion time scales, flamelet curvature, strong variations of the scalar dissipation within the flamelet structure require more detailed analysis.

**Strain Rate and Scalar Dissipation**

The effect of strain rate on the structure of the laminar flame has been described as follows for the H₂/Air system (Dixon and Missaghi 1988). With increasing strain rate the flame becomes thinner and the volumetric heat release rate increases right up to the extinction limit. The higher reaction rates are forced on the system by the reduced residence times, and until extinction occurs they are accompanied by lower maximum temperatures, reaction product concentrations, and increased breakthrough of reactants beyond the stoichiometric position. In common with the volumetric heat release rate, net radical production rates are comparatively low in the "near-equilibrium" flames with low strain rates, and they increase with increasing strain rates.

A characteristic strain rate for an opposed-flow diffusion flamelet can be defined as \( a_s = \frac{v}{2d} \), where \( v \) is the speed of the fuel and oxidizer jets, and \( d \) is the distance between the jet nozzles. Instead of using the strain rate to quantify the departure from equilibrium, it is expedient to use the scalar dissipation, denoted by \( \chi \). The instantaneous scalar dissipation is defined as \( \chi = 2D|\nabla f|^2 \), where \( D \) is a representative diffusion coefficient. Scalar dissipation, \( \chi \), varies along the axis of the flamelet. For the counterflow
geometry, the flamelet strain rate $a_s$ can be related to the scalar dissipation at the position where $f$ is stoichiometric by Peters (1984) and Bray and Peters (1994):

$$x_{st} = \frac{a_s \exp \left( -2[erfc^{-1}(2f_{st})]^2 \right)}{\pi}$$  \hspace{1cm} (2.62)

where $a_s$ is the characteristic strain rate, $f_{st}$ is stoichiometric mixture fraction and $erfc^{-1}$ is the inverse complementary error function.

The instantaneous scalar dissipation rate at stoichiometric conditions, $\chi_{st}$, is used as the essential nonequilibrium parameter in the flamelet modeling. It has the dimensions 1/s and may be interpreted as the inverse of characteristic diffusion time. $\chi$ can be calculated at each location in the flow field. $x_{st}$ will increase due to stretching, and in the limit $\chi_{st} \to 0$ the chemical equilibrium assumption applies. Local quenching of the flamelet occurs when $x_{st}$ exceeds a critical value. For a turbulent flame, the scalar dissipation rate can be calculated from the basic turbulence quantities and mixture fraction variance for the flow field as Bray and Peters (1994):

$$x_{st} = C_x \varepsilon f'^2$$  \hspace{1cm} (2.63)

where $C_x$ is a constant with a value of 2.0 (Liew et al. 1984), $k$ is the turbulence kinetic energy, $\varepsilon$ is turbulence dissipation rate and $f'^2$ is the mixture fraction variance. The dimensionless scalar dissipation is defined as:

$$\chi_{st,d} = \frac{\chi_{st}}{\chi_{st,max}}$$  \hspace{1cm} (2.64)

As mentioned above, the thermo-chemical state in the turbulent non-premixed flame is represented by an ensemble of laminar diffusion flamelets. Most of the studies use a counter flow diffusion flame to obtain flamelet data, conveniently known as flamelet library.
Figure 2.3: Tsuji counterflow diffusion flame geometry

Figure 2.4: Counterflow diffusion flame planar geometry
Alternatively, flamelet equations on the conserved scalar space can be used to generate flamelet library (Lentini 1994; Mauss et al. 1990). In the present study, both methods are used. A brief description of both methods for generating flamelet library is presented in the following subsections.

Counterflow diffusion flame

The counterflow geometry is very often used in experimental and numerical studies of diffusion flames because it leads to an essentially one-dimensional diffusion flames structure. Figure 2.3 and 2.4 show flames that have been established between an oxidizer stream and a fuel stream from opposite directions for both axisymmetric and planar geometry. There exists exact solution in terms of a similarity coordinate, (Dixon-Lewis et al. 1984 or Peters and Kee, 1987). N. Peters (2000) also derived equations using the y coordinate directly by introducing

\[ u = Ux, \]

The velocity in the y direction is denoted by \( v \) and the gradient of the velocity \( u \) in the x direction by \( U \). Then the governing equations are:

**Continuity:**

\[ \frac{d(\rho v)}{dy} + (j + 1)\rho U = 0, \quad (2.65) \]

**Momentum:**

\[ \rho v \frac{dU}{dy} = -\rho U^2 + P + \frac{d}{dy} \left( \mu \frac{dU}{dy} \right), \quad (2.66) \]

**Mixture fraction:**

\[ \rho v \frac{dZ}{dy} = \frac{d}{dy} \left( \rho D \frac{dZ}{dy} \right), \quad (2.67) \]

**Reactive scalars:**

\[ \rho v \frac{dy_i}{dy} = \frac{d}{dy} \left( \rho D_i \frac{dy_i}{dy} \right) + \omega_i, \quad (2.68) \]
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Here $j$ is a geometrical parameter and $j = 0$ applies for planar and $j = 1$ for the axisymmetric configuration. The parameter $P$ represents the axial pressure gradient and is related to the strain rate by $P = \rho_0 a^2$.

Flamelet library using Tsuji burner geometry

The tsuji burner consists of a laminar fuel flow issuing from a porous cylinder of radius $R$ and a laminar air stream in the opposed direction as shown in Fig. 2.3. A stable reaction zone is established in the forward stagnation region of the cylinder. The fuel and oxidizer diffuse toward the reaction zone with velocities proportional to the concentration gradients. There is a strong coupling between the fluid mechanics (fuel-air mixing rate) and chemical kinetics (reaction rates) leading to the reaction zone structures which are strong functions of the velocity gradients.

The flow in the stagnation region is given by (Drake and Blint 1988; Peters 1984):

\begin{align*}
u &= ax \quad (2.69) \\
v &= -ay \quad (2.70)
\end{align*}

where $x$ and $y$ are the coordinates parallel and perpendicular to the burner surface respectively, $u$ and $v$ are the corresponding velocity components and $a$ is the velocity gradient. Dimensionless velocities $f'$ and $V$ together with a density weighted $y$ coordinate are often introduced as:

\begin{align*}
f' &= \frac{u}{u_e} \quad (2.71) \\
V &= \frac{\rho v}{\rho \mu_e a} \quad (2.72)
\end{align*}

Where $u_e$ is the value of $u$ at $y = \infty$. 

The non-dimensional distance \( \eta \) may be expressed as:

\[
\eta = \sqrt{\frac{a}{\rho_e \mu_e}} \int_0^1 \rho dy \quad (2.73)
\]

Since there is no tangential fuel velocity, \( f' = 0 \) at the burner wall. Since \( u \to u_e \) as \( y \to \infty \), \( f' = 1 \) in the air stream. Along the stagnation point streamline direction (\( x=0 \)), the following differential equations apply:

Continuity:

\[
f' + \frac{dV}{d\eta} = 0 \quad (2.74)
\]

Momentum:

\[
(f')^2 + V \frac{dV}{d\eta} - \frac{\rho}{\rho_e} = \frac{d}{d\eta} \left( C \frac{df''}{d\eta} \right) \quad (2.75)
\]

where \( C = \frac{\rho H}{\rho_e \mu_e} \)

Species:

\[
V \frac{dY_\alpha}{d\eta} - \frac{\omega_\alpha W_\alpha}{\rho a} + \frac{1}{\sqrt{\rho e \mu e a}} \frac{d}{d\eta} \left( \rho Y_\alpha V a \eta \right) = 0 \quad \text{for } \alpha = 1, 2, \ldots, N \quad (2.76)
\]

Energy:

\[
V \frac{dT}{d\eta} = \frac{1}{\sqrt{\rho e \mu e a}} \frac{d}{d\eta} \left( \frac{\rho \frac{dT}{d\eta}}{c_p} \right) - \frac{1}{c_p} \sum_{\alpha=1}^{N} \rho Y_\alpha V a \eta C p a \frac{dT}{d\eta} \quad (2.77)
\]

In these equations, \( Y_\alpha \) and \( W_\alpha \) are the mass fraction and molecular weight of species \( \alpha \), and \( T, C_p \) and \( \rho \) are the temperature, specific heat and density of the mixture. \( \omega_\alpha \) is the diffusion co-efficient of the \( \alpha \)-th species in the \( \eta \) direction. The thermal diffusion is
neglected in the calculation. The chemical production rate of the $\alpha$-th species per unit volume, $\dot{\omega}_\alpha$, is calculated from:

$$\dot{\omega}_\alpha = \sum_{\beta=1}^{R} \dot{\omega}_{\alpha\beta}$$  \hspace{1cm} (2.78)

where $\dot{\omega}_{\alpha\beta}$ is the reaction rate of $\alpha$-th species in the $\beta$-th elementary reaction step. The reaction mechanism used in the present study is taken from (Peters 1993) and is given in appendix A.

Flamelet generation with equations in the mixture fraction space

The laminar counterflow diffusion flame equations can be transformed from physical space (with $x$ as the independent variable) to mixture fraction space (with $f$ as the independent variable) Pitsch and Peters (1998). The field equation for the mixture fraction that determines the location of the flame surface is:

$$\rho \frac{\partial f}{\partial t} + \rho v \cdot \nabla f = \nabla \cdot (\rho D \nabla f)$$  \hspace{1cm} (2.79)

Then a simplified set of the mixture fraction space equations are solved (Pitsch, Barths and Peters 1996). Here, $N$ equations are solved for the species mass fractions, $Y_i$.

The flamelet equations are devised, based on universal co-ordinate transformation on the mixture fraction space, in the following form (Peters 1984; Bray and Peters 1994):

$$\rho \frac{\partial \psi_i}{\partial t} = \rho \frac{K}{2} \frac{\partial^2 \psi_i}{\partial Z^2} + \omega_i$$  \hspace{1cm} (2.80)

$$\rho \frac{\partial T}{\partial t} = \rho \frac{K}{2} \frac{\partial^2 T}{\partial Z^2} - \sum_{i=1}^{n} \frac{h_i}{c_p} \omega_i$$  \hspace{1cm} (2.81)
where $\chi$ is the scalar dissipation rate and has to be specified as an input. This formulation has widely been used to generate flamelet library (Mauss et al. 1990; Seshadri et al. 1990; Lentini 1994).

This formulation has the advantage that it is independent of the flow field and the scalar dissipation rate, $\chi$ implicitly incorporates the convection and diffusion effects normal to the surface of the stoichiometric mixture (Bray and Peters 1994). However, the formulation is derived by neglecting the higher order terms involving convection and curvature along the stoichiometric mixture fraction surface and also based on the assumption of unity Lewis number. The effects of higher order terms are sometimes important (Bray and Peters 1994). The assumption of unity Lewis number also influences the flamelet structure specially in the rich zone (Pitch and Peters 1998). An advanced flamelet formulation which includes the differential diffusion effects has been reported by Pitch and Peters (1998). This formulation is quite complex and some brief ideas about it have been given below.

**Differential Diffusion and Extinction Limit**

The limiting behaviour of the flamelet solutions as the scalar dissipation $\chi \to 0$ is an important issue because $\chi \to 0$ implies the flamelet domain length, $L \to \infty$. As the domain length increased, mixing and reaction rates become slower, and the chemical state moves closer to equilibrium. At some point, however, the length of the flamelet domain will become greater than the physical dimension of the combustor. The differential diffusion effects are generally negligible at the larger scales because of turbulent transport. Thus, as the flamelet domain becomes longer, turbulence contributes increasingly to species transport inside the flamelet and the effects of differential diffusion diminish. Furthermore the correct limiting behaviour to equilibrium chemistry will only be obtained when differential diffusion is absent.

As differential diffusion is predominantly small-scale phenomenon, it is difficult for a single point description to capture this effect. An important question arises that how differential diffusion scales with Reynolds number. The argument is often made that at high Reynolds numbers, molecular effects are confined to high wavenumbers and thus have a small effect on single-point quantities of interest but it appears that chemical
reactions increase the magnitude of the effect. This is most probably due to an increase in scalar gradients that accompanies depletion of the reactant species.

One of the advantages of flamelet models is their ability to easily incorporate the effects of complex mass diffusion as well as complex chemical kinetics. Since the flamelet model provides a spatial variation to the flame structure, the effects of differential transport of species can be combined with the effects of differential reaction rates to compute a detailed, complex flame structure. Thus the conclusion to be inferred is that accurate treatment of differential diffusion can be just as important as accurate modeling of chemical kinetics.

Another limiting case to consider is the behaviour of the flamelet solutions as $\chi \to \infty$ ($L \to 0$). As the dissipation rate is increased, both the mixing and reaction rate increase, while the maximum flame temperature gradually decreases until the flame temperature becomes so low that reaction rates cannot increase any further due to the effects of Arrhenius kinetics. Once this critical turning point is reached ($\chi = \chi_{\text{crit}}$), further increase in $\chi$ will cause the flame to extinguish.

Statistics of non-equilibrium parameter

In the laminar flamelet model, the thermo-chemical composition of the turbulent flame is completely determined by two parameters, the mixture fraction and a non-equilibrium parameter, the scalar dissipation rate. In turbulent flow fields, these parameters are statistically distributed. It is therefore necessary to know the statistical distribution of the mixture fraction and the scalar dissipation rate. In virtually all laminar flamelet model formulations, it is assumed that the mixture fraction and the scalar dissipation rate are statistically independent. This assumptions drastically simplifies the formulations. Thus, the average value of scalar variables in a turbulent flow field is given by:

$$\bar{\phi} = \int_{0}^{\infty} \int_{0}^{1} \phi(Z; \chi) P(Z) P(\chi) dZ d\chi$$

(2.82)

The pdf for mixture fraction is assumed to be a beta function (Peters 1984). The pdf of scalar dissipation rate is assumed to be a log-normal function and experimental
evidence justifies this assumption (Effelsberg 1988). The mean scalar dissipation rate in the turbulent flow field is modelled according to:

$$
\overline{\chi} = C_\chi \frac{\overline{\varepsilon}}{k} Z^2
$$

(2.83)

where $C_\chi$ is a constant with a value of 2.0 (Liew et al. 1984). The width of the log-normal distribution is given in terms of variance, $\sigma$ (Peters 1984). The value of $\sigma$ is assigned as 2.0 after Peters (1984).

In principle, the local and global extinction of turbulent flames can be described by the distribution of the scalar dissipation rate. Local extinction events occur when the scalar dissipation rate exceeds the extinction limit $\chi_q$ and, since the scalar dissipation rate is distributed in turbulent field, the probability of extinction is given by:

$$
P_{\text{ext}} = \int_{\chi_q}^{\infty} P(\chi) d\chi
$$

(2.84)

The scalar dissipation rate, and consequently the probability of extinction, depends on the flow time scales, which vary across the turbulent flame. For higher mixing rate, the value of the scalar dissipation rate is high and the pdf, $P(\chi)$, shifts towards the higher values of the scalar dissipation rate. This results in a higher probability of extinction and the presence of a larger portion of extinguished flamelets. Percolation theory (Peters 1984; Peters 1986) predicts that when the probability of extinction exceeds a certain critical value $P_c$, the flame will no longer be interconnected and global extinction will occur. In that case, the flamelets representing the thermo-chemical state of a turbulent flame at a location in question are all extinguished. This approach has been used to analyse the lift-off phenomenon of turbulent jet diffusion flames (Sandlers and Lamers 1994). In the region near the burner nozzle, the intense turbulent mixing leads to a higher scalar dissipation rate; if $P_{\text{ext}}$ exceeds $P_c$, global extinction occurs and the flame is lifted. Further downstream, where the scalar dissipation rate decreases to such a value that $P_{\text{ext}} = P_c$, the flame is stabilised.
Though the flamelet model has been successful in the predicting lift-off phenomenon, it is not yet clear whether the same formulation will be successful in predicting the local extinction phenomenon, often observed in turbulent CH\textsubscript{4} flames (Masri et al. 1996). In the present study, the laminar flamelet model is assessed for a number of fuel mixtures as well as for different Reynolds numbers to investigate its suitability for flames both near and far away from chemical equilibrium.

Non-equilibrium parameter

Two parameters are currently in use to describe the non-equilibrium effects: the scalar dissipation rate and the strain rate. The scalar dissipation rate is theoretically the correct parameter and should account for non-equilibrium effects caused by both convection and diffusion (Bray and Peters 1994). However, recent studies suggest that the use of strain rate is more appropriate (Sandlers and Lamers 1994; Bray and Peters 1994; Bradley et al. 1990; Fairweather et al. 1991). The arguments for using the strain rate are given in Bray and Peters (1994). The scalar dissipation rate, given by equation (2.83), depends on two physical parameters: (1) the turbulence time scale $\frac{\varepsilon}{k}$ and (2) the mixture fraction variance, $Z^2$. According to Bray and Peters (1994), the non-equilibrium parameter used to couple the thermo-chemical state of the turbulent flame to that in the library should depend only on the turbulent time scale, represented by $\frac{\varepsilon}{k}$. The mixture fraction fluctuations should not influence the chemistry as long as these do not enter into the reaction zone. This differentiation is very important for partially premixed turbulent flames where the partial mixing of fuel and air occurs before chemical reaction takes place. The flamelets in a turbulent premixed flame do not extend from pure fuel ($Z=1$) to pure air ($Z=0$), rather they span over a smaller part of mixture fraction space. Corresponding partially premixed flamelets can be constructed by moving the boundary conditions towards the reaction zone while keeping the strain rate and consequently the scalar dissipation rate constant at the reaction zone. The structure of partially premixed flamelets should depend on the temperature and the concentrations imposed on the new boundary. However, for fuels with a small stoichiometric mixture fraction such as hydrogen or methane, the flamelet profiles are
not expected to be influenced by partial premixing of the fuel stream. This is because the reaction zone is still far away from the fuel boundary. However, the corresponding reduction of the mixture fraction variance across the flamelet would decrease the mean scalar dissipation rate (see Eq. 2.83), and subsequently will influence the thermo-chemical state of the turbulent flame, if the scalar dissipation rate is used as the non-equilibrium parameter. Based on above analysis, Bray and Peters (1994) have proposed to use the strain rate as the non-equilibrium parameter and to relate its mean to turbulent time scales as:

\[ \bar{a} = \frac{\bar{\varepsilon}}{k} \]  

(2.85)

Correspondingly, a new scalar dissipation rate, producing an equivalent coupling between the time scales of the turbulent flame and the library, can be defined according to Bray and Peters (1994). The expression for the modified scalar dissipation rate is given by:

\[ \chi = C \frac{\bar{\varepsilon}}{k} (\Delta Z)^2 \]  

(2.86)

The scalar dissipation rate defined by the above expression differs from the standard scalar dissipation rate defined by the Eq. (2.62) in the mixture fraction variance term. The new relation is based on a fixed mixture fraction variance, \( \Delta Z \), rather than on the actual mixture fraction variance, \( Z^* \), which varies along the turbulent flame. \( \Delta Z \) is the thickness of the laminar flamelets in the mixture fraction space and it contains the reaction zone and surrounding diffusive layers. For fuel with small stoichiometric mixture fraction, \( \Delta Z \) can be taken as \( \Delta Z = 2Z_{st} \) as a first approximation (Bray and Peters 1994). The use of the strain rate as well as the new definition of the scalar dissipation rate has been very successful in predicting the lift-off phenomenon (Sandlers and Lamers 1994; Bray and Peters 1994). However, the flamelet model using the strain rate as the non-equilibrium parameter has failed to predict the scaling behavior of NO\(_x\) (Sanders and Gökalp 1995; Schlatter et al. 1996). Sanders et al. (1997) have investigated the effects of different non-equilibrium parameter on the NO\(_x\) scaling.
behavior by using the strain rate, the standard scalar dissipation rate defined by Eq. (2.62) and the modified scalar dissipation rate defined by Eq. (2.86). They found that only the use of the standard scalar dissipation rate, defined by Eq. (2.62), as the non-equilibrium parameter, can produce the correct trend of NO\textsubscript{x} scaling. Thus, the question of the correct parameter for accounting the non-equilibrium effect is still open and it is further investigated in the present study.

### 2.6.3 Eddy break-up model

Devised by Spalding (1971), this model is based on phenomenological analysis of turbulent combustion assuming high Reynolds ($Re \gg 1$) and Damköhler ($Da \gg 1$) numbers. The reaction zone is viewed as a collection of fresh and burnt gases pockets. Eddy-Break-Up (EBU) modelling approach is very different from the modelling approaches described so far specially with respect to chemistry modelling and the turbulence-chemistry interaction modelling. The eddy break-up model is an empirical model for expressing the mean chemical reaction rate in the transport equations of the mean mass fractions. Spalding (1971) observed that the reaction rate in a premixed flame is only weakly dependent on temperature, pressure and mixing strength, instead the reaction process is controlled by turbulence mixing. Based on these observations, he proposed a model for the reaction rate, which did not account explicitly the effects of chemical kinetics. This model is known as the eddy break-up model. According to the model, the mean reaction rate is proportional to the rate at which larger eddies are broken into smaller eddies. The expression for the mean reaction rate of fuel according, to this model, is given by:

\[
\bar{\omega}_f = C_{EBU} \frac{\bar{\varepsilon}}{k} (\bar{Y}_f^p)^{1/2}
\]  

(2.87)

where $\bar{Y}_f^p$ is the fluctuation of fuel mass fraction and $C_{EBU}$ is the model constant. This model faces difficulties in determining the fluctuations of fuel concentrations. Models similar to the original eddy break-up model have been proposed by a number of researchers. The most popular of these models is the one proposed by Magnussen and Hjertager (1976). The expression for the mean reaction rate in the eddy break-up model
proposed by Magnussen and Hjertager (1976), which is popularly known as Magnussen's model, is given by:

\[
\bar{\omega}_f = \frac{\bar{\varepsilon}}{k} \min \left( A\bar{Y}_{fu}, A\frac{\bar{Y}_{ox}}{s}, B\frac{\bar{Y}_{pr}}{1+s} \right)
\]

(2.88)

where \(Y_{fu}, Y_{ox}\) and \(Y_{pr}\) are the concentration of fuel, oxygen and products respectively, \(s\) is the stoichiometric oxygen fuel ratio. \(A\) and \(B\) are model constants with values of 4.0 and 2.0 respectively. The crucial difference between this model and the original eddy break-up model is that this model relates the dissipation of eddies with the mean concentrations of intermittent quantities instead of the fluctuating concentrations. This model also recognises that the dissipation of eddies containing different species can be rate determining for different conditions. Magnussen's eddy break-up model has been widely used for calculations of industrial flows and is also implemented and evaluated in the present study. An extended version of the model known as the eddy dissipation concept (EDC) model is also available which can further include finite rate chemistry effects (Gran 1994). Several options are available in the numerical application of the EBU model. In one method, as utilised in Magnussen and Hjertager (1976), the transport equation for the fuel mass fraction is solved with the rate of reaction available from the EBU model. An additional equation of mixture fraction is solved from where remaining species are calculated. The second method, as used in the commercial code FLUENT, is to solve the transport equations of all the species involved in the combustion process. The EBU model gives the source term for fuel and the source terms for the remaining species are calculated from the reaction rate expression with an appropriate stoichiometric relation. The second method is implemented in the present study. In this method, the following transport equations are solved for mass fraction of fuel, oxygen, product and enthalpy:

Mass fraction of fuel:

\[
\frac{\partial}{\partial x_j} (\bar{\rho} \bar{u}_j \bar{Y}_{fu}) = \frac{\partial}{\partial x_j} \left( \frac{\mu_l}{\sigma_t} \frac{\partial \bar{Y}_{fu}}{\partial x_j} \right) - \bar{\rho} \bar{\omega}_f
\]

(2.89)
Chapter 2. Mathematical Models

Mass fraction of oxygen:
\[ \frac{\partial}{\partial x_j} (\overline{\rho u_j} \overline{Y_{ox}}) = \frac{\partial}{\partial x_j} \left( \frac{\mu_t}{\sigma_t} \frac{\partial \overline{Y_{ox}}}{\partial x_j} \right) - s \overline{\rho \omega_f} \]  
(2.90)

Mass fraction of product:
\[ \frac{\partial}{\partial x_j} (\overline{\rho u_j} \overline{Y_{pr}}) = \frac{\partial}{\partial x_j} \left( \frac{\mu_t}{\sigma_t} \frac{\partial \overline{Y_{pr}}}{\partial x_j} \right) - (1 + s) \overline{\rho \omega_f} \]  
(2.91)

Enthalpy:
\[ \frac{\partial}{\partial x_j} (\overline{\rho u_j} \tilde{h}) = \frac{\partial}{\partial x_j} \left( \frac{\mu_t}{\sigma_t} \frac{\partial \tilde{h}}{\partial x_j} \right) \]  
(2.92)

where, \( Y_{fu}, Y_{ox}, \) and \( Y_{pr} \) are mass fraction of fuel, oxygen and product respectively; \( s \) is the stoichiometric mass of oxygen, and the fuel rate of fuel burning, \( \overline{\omega_f} \), is given by Eq.(2.88). The mean temperature can be obtained from:
\[ \tilde{h} = \sum_{\alpha=1}^{N} \tilde{Y}_\alpha h_\alpha (\tilde{T}) \]  
(2.93)

and the mean density is determined from the equation of state for a mixture of perfect gases:
\[ \overline{\rho} = \frac{\overline{P}}{\tilde{T} R \sum_{\alpha=1}^{N} \frac{\tilde{Y}_\alpha}{M_\alpha}} \]  
(2.94)

The refinement of the "eddy-break-up" model involved regarding a turbulent burning mixture as comprising inter-mingled fragments of just two gases, namely:
- a completely unburned mixture of fuel and oxidant; and
- the completely burned products of its combustion.

It was recognised that, at the interfaces between the two sets of fragments, thin layers of gas in various stages of incomplete combustion must exist; but the fraction of the total
volume occupied by these gases was regarded as much less than unity; The only important consequence of their existence was the primary chemical transformation

\[
\text{Fuel + Air} \rightarrow \text{Products}
\]

which took place in them.

The rate of that transformation, per unit volume of the total space, was treated as being governed, however, by the rate of turbulent micro-mixing, for which the quantity, \( \varepsilon/k \) was a convenient measure.

The EBU model was found attractive because the reaction rate is simply written as a function of known quantities without any additional transport equation and is available in most of the commercial CFD codes. The modelled reaction rate does not depend on the chemical characteristics and assumes a homogeneous and isotropic turbulence. Eddy-Break-Up modelling tends to overestimate the reaction rate, especially in highly strained regions, where the ratio \( \varepsilon/k \) is large (flame-holder wakes, walls, etc.).
CHAPTER 3

Numerical Implementation

The numerical solution of heat transfer, fluid flow, combustion and other related processes is possible when the laws governing these processes have been expressed in mathematical form, generally in terms of differential equations. This chapter summarises the numerical methods employed to implement the various mathematical models presented in the previous chapter. The numerical methods are of crucial importance in the detailed study of combustion processes. A reliable and accurate numerical method is needed so that the errors associated with the numerical procedures do not prevent the evaluation of the underlying mathematical models. There are a large number of numerical methods available in the literature for solving the governing equations of flow, heat transfer and combustion (Patankar 1980; Versteeg and Malalasekera 1995; Ferziger and Peric 1996). A brief description of the widely used numerical methods is given in the following sections. The solution method for the flow field calculations is described without any reference to combustion models. The incorporation of the combustion model into the general flow calculations is described in the later sections. It is not been possible to cover even a substantial fraction of the models here and a short overview of some methods has been given.

3.1 Conservation equations

All conservation equations of relevance here have similar structure and possess a common form. For this reason a single general conservation equation is used to demonstrate discretisation methods for the terms, which are common to all conservation equations (convection, diffusion, and sources). The heat transfer, fluid flow, combustion and other related processes can be described by the governing equations of
conservation of mass, momentum, turbulence kinetic energy, energy dissipation rate, mixture fraction and mixture fraction variance equations. The time-independent conservation and transport equations can be written in the following generalised form:

\[
\frac{\partial}{\partial x_j} (\bar{\rho} \bar{u}_j \phi) = \frac{\partial}{\partial x_j} \left( \Gamma_\phi \frac{\partial \phi}{\partial x_j} \right) + S_\phi
\]  

(3.1)

where \( \phi \) is the general dependent variable. These equations differ only in the form of the source term, \( S_\phi \) and their diffusive transport coefficient, \( \Gamma_\phi \). The recognition that all the relevant conservation equations for heat and mass transfer, fluid flow, combustion, turbulence and other related phenomena can be thought of as particular cases of the general \( \phi \) equation is an important time-saving step. As a consequence, we need to concern ourselves with the numerical solution of only Eq. (3.1). In the construction of computer program, it is sufficient to write a general sequence of instructions for solving Eq. (3.1), which can be repeatedly used for different meanings of \( \phi \) along with appropriate expressions for \( \Gamma_\phi, S_\phi \) and with the relevant initial and boundary conditions.

Thus, along with the appropriate initial and boundary conditions, these elliptic equations are solved numerically by a standard finite volume method. The detailed derivation and formulation of the finite volume method can be found in (Patankar 1980; Versteeg and Malalasekera 1995). In this study, only a brief description of the general make-up of the methods has been provided.

### 3.2 Discretisation of equations

The set of algebraic equations involving the unknown values of dependent variable at chosen grid points, which is known as discretised equations or difference equations, are derived from the differential equation governing the dependent variable. The differential equations are discretised over small control volumes or cells to obtain the difference equations. The calculation domain is divided into a finite number of such control volumes to form a computational grid system. Numerical values of the scalar variables and pressure are determined at the intersection of these mesh lines (grid
nodes). The velocities are calculated at a point mid-way between these grid nodes. In
the estimation of velocity components, there is a significant benefit to be obtained by
arranging them on grids that are different from the grid used for all other variables.

Such a displaced or "staggered" grid for velocity component was first used by Harlow
and Welch (1965) in their MAC method and later used in other methods developed by
Harlow et al. It forms the basis of the SIMPLE procedure of Patankar and Spalding
(1972). This "staggered" location for velocities makes the calculation of the pressure
gradients easy and necessitates the adoption of different control volumes for each of the
velocity component. Figure 3.1 shows such control volumes for a 2-D case in Cartesian
co-ordinates. The descretisation of Eq. (3.1) over the corresponding control volume for
the variable in question together with appropriate assumptions about the way in which
the quantities vary between grid nodes leads to algebraic equations of the following
form:

$$ a_p \phi_p = \sum_{i=E,W,N,S} a_i \phi_i + S_c \tag{3.2} $$

where, $P$ is the central node and $E, W, N, S$ are the neighbour nodes of the control
volume. In the integration procedure, the source term, $S_\phi$, in Eq.(3.1) is linearised as:

$$ S_\phi = S_p \phi_p + S_c \tag{3.3} $$

where, $S_p$ is the coefficient of $\phi_p$ and $S_c$ stands for the constant part of $S_\phi$. According to
the negative-slope linearization of source term rule (Patankar 1980), it is required that
$S_p$ will not be positive. Thus, the coefficient $S_p$ must always be less than or equal to
zero. The coefficient of the central node is given by:

$$ a_p = \sum a_i - S_p \tag{3.4} $$

The neighbouring coefficients $a_i$ contain the influence of the convective and diffusive
fluxes through the cell faces. The ratio of convective to diffusive flux at a given cell
face is conventionally known as a cell Peclet number e.g. for a face with surface normal
to the x-direction, the cell Peclet number is defined as:

$$ Pe_{\phi,x} = \frac{\rho u \Delta x}{\Gamma_{\phi}} \tag{3.5} $$
Chapter 3. Numerical Implementation

3.2.1 Discretisation schemes

The convection term has an inseparable connection with the diffusion term, and therefore, need to be handled both as one unit. The integration of the convective terms on the left hand side of Eq. (3.1) leads to the need to interpolate the \( \phi \) values at the cell faces from the adjacent nodal values. This interpolation practice constitutes the most problematic element of the discretisation scheme (the diffusive fluxes are always discretised using second order accurate central differencing).

The usual approach is to define control volumes (CVs) by a suitable grid and assign the computational node to the CV centre. However, one could as well define the nodal locations first and construct CVs around them, so that CV faces lie midway between nodes. Fig. 3.2 shows a two dimensional control volume and associated nodal stencil.
The advantage of the second approach is that central difference approximations of derivatives at CV faces are more accurate when the face is midway between two nodes. The discretisation of the convection terms with the central differencing scheme (CDS) produces oscillations, which are physically unrealistic (Patankar 1980). The hybrid scheme (combination of upwind differencing scheme and CDS) eliminates these oscillations, however the scheme has known tendency to introduce numerical diffusion because of switching to the first order upwind scheme for \(|\text{Pe}|>2\). The numerical diffusion is specially prominent in the presence of skewness (i.e. when the flow is not aligned to the grids) Patankar (1980). Minimisation of the artificial diffusion by refining grids incurs unacceptable computational penalty. The higher order schemes, such as the third-order upstream weighted QUICK (Quadratic Upwind Interpolation for Convective Kinematics) scheme of Leonard (1979) may overcome this drawback. Although the higher order discretisation schemes avoid the problems associated with first-order upwinding, they are more unstable and do not necessarily improve the solutions. For instance, during a comparative study of different discretisation schemes, Lien and

**Figure 3.2:** Two dimensional control volume and associated nodal stencil.
Leschziner (1994) and Alvarez and Jones (1993) confirmed that the second-order TVD scheme and the third-order QUICK scheme yield identical solution and the TVD scheme is much more stable and computationally efficient. In any case, higher order schemes have little influence in computations involving turbulence models, such as the $k-\varepsilon$ model which are generally dominated by the production and destruction terms rather than the convection term (Lien and Leschziner 1994; Alvarez and Jones 1993).

The modelling of the turbulent reacting flows is subjected to a number of potential sources of error. These include those arising from the turbulence model, those arising from the combustion model and those, which results from discretisation of the resulting set of differential equations. It is clearly of the utmost importance to isolate the error associated with the discretisation scheme in order to draw definitive conclusions on the performance of calculation methods. In order to investigate the numerical error associated with the discretisation scheme three different schemes are used in this study, which are:

**Hybrid:** The hybrid scheme is a combination of central differencing and upwindschemes. It is identical with the central-difference scheme for the Peclet number range $|Pe| \leq 2$ and outside this rage it reduces to the upwind scheme in which the diffusion has been set equal to zero. The name hybrid is indicative of a combination of the central-difference and upwind schemes, but it is best to consider it as the three-line approximation to the exact curve (Patankar 1980). This scheme produces a realistic solution and is highly stable. The hybrid scheme has been widely used for predicting practical flows. The disadvantage is that this scheme is first order accurate and produces false diffusion.

**Power law:** The power law differencing scheme of Patankar (1980) is a curve fit to the exact solution of a one dimensional, steady state, convection-diffusion flow problem. In this scheme, the diffusion is set to zero when the cell Pe exceeds 10. If $0<Pe<10$, the flux is evaluated by using a polynomial expression. For $|Pe| > 10$, the power-law scheme becomes identical with hybrid scheme. This scheme produces better result than the hybrid scheme, but it also suffers from the problem of false diffusion.
TVD: The total variation diminishing (TVD) scheme, first devised for gas dynamics problems, has also been used in the turbulent combusting flows (Jones 1994). The TVD scheme is stable, second order accurate and is easy to implement. In this scheme, the cell face values needed in the differencing of the convection term are written as upwind values plus correction terms which overall lead to second order accuracy, but where the correction is limited to prevent spatial oscillations by means of a limiter. There are a wide range of limiters available in the literature (Van Leer 1974; Chakravarthy and Osher 1983; Lien and Leschziner 1994). In the present study, Van Leer's (Van Leer 1974) and UMIST (Upstream Monotonic Interpolation for Scalar Transport) Lien and Leschziner (1994) limiters are employed. In the TVD scheme, the upwind contribution is included in the neighbouring coefficients, while the anti diffusive flux is implemented via a "deferred correction" source term $S_{\phi}^{DC}$.

The coefficients of the discretised Eq. (3.2) that result from applying the above mentioned schemes are given in Table (3.1).
### Table 3.1: Coefficients for different scheme

<table>
<thead>
<tr>
<th>Hybrid scheme</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a_E = \max[-F_e, (D_e - F_e/2), 0]$</td>
</tr>
<tr>
<td>$a_W = \max[F_w, (D_w + F_w/2), 0]$</td>
</tr>
<tr>
<td>$a_N = \max[-F_n, (D_n - F_n/2), 0]$</td>
</tr>
<tr>
<td>$a_S = \max[F_s, (D_s + F_s/2), 0]$</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Power law scheme</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a_E = D_e \max[0, (1-0.1</td>
</tr>
<tr>
<td>$a_W = D_w \max[0, (1-0.1</td>
</tr>
<tr>
<td>$a_N = D_n \max[0, (1-0.1</td>
</tr>
<tr>
<td>$a_S = D_s \max[0, (1-0.1</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>TVD scheme</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a_E = D_e + \max(-F_e, 0)$</td>
</tr>
<tr>
<td>$a_W = D_w + \max(F_w, 0)$</td>
</tr>
<tr>
<td>$a_N = D_n + \max(-F_n, 0)$</td>
</tr>
<tr>
<td>$a_S = D_s + \max(F_s, 0)$</td>
</tr>
</tbody>
</table>

\[ S_{\Phi}^{DC} = \frac{[F_e^{-} \Psi(r_e^{-})/2 - F_e^{+} \Psi(r_e^{+})/2](\Phi_E - \Phi_P)}{2} + [F_w^{-} \Psi(r_w^{-})/2 - F_w^{+} \Psi(r_w^{+})/2](\Phi_P - \Phi_W) + [F_n^{-} \Psi(r_n^{-})/2 - F_n^{+} \Psi(r_n^{+})/2](\Phi_N - \Phi_P) + [F_s^{-} \Psi(r_s^{-})/2 - F_s^{+} \Psi(r_s^{+})/2](\Phi_P - \Phi_W) \]

where, \( F^+ = \frac{F + |F|}{2} \quad F^- = \frac{F - |F|}{2} \)

\( r_e^+ = \frac{\Phi_P - \Phi_W}{\Phi_E - \Phi_P} \quad r_e^- = \frac{\Phi_E - \Phi_{EE}}{\Phi_p - \Phi_E} \quad r_w^+ = \frac{\Phi_W - \Phi_W}{\Phi_P - \Phi_W} \quad r_w^- = \frac{\Phi_P - \Phi_E}{\Phi_W - \Phi_P} \)

and \( r_n^+ = \frac{\Phi_P - \Phi_N}{\Phi_N - \Phi_{NN}} \quad r_n^- = \frac{\Phi_N - \Phi_N}{\Phi_p - \Phi_N} \quad r_s^+ = \frac{\Phi_S - \Phi_{SS}}{\Phi_P - \Phi_S} \quad r_s^- = \frac{\Phi_P - \Phi_N}{\Phi_S - \Phi_P} \)

and limiters are defined as

\( \Psi(r) = \frac{r^+ + |r|}{1 + |r|} \) for Van Leer limiter

\( \Psi(r) = \max[0, \min(2r, 0.25 + 0.75r, 0.75 + 0.25r, 2)] \) for UMIST limiter

In the expressions, \( F = \rho u \quad D = \frac{\Gamma}{\delta x} \quad Pe = \frac{F}{D} \)
3.3 Boundary conditions

The discretisation of transport equations over CVs provide algebraic equations and each CV provides one algebraic equation. Volume integrals are calculated for every CV, but fluxes through CV faces coinciding with the domain boundary require special treatment. The transport equations of the scalar variables are of elliptic type in most of the cases (Patankar 1980). The solution of elliptic partial differential equations requires information to be provided at all points on a closed boundary surrounding the solution domain. These boundary fluxes must either be known, or be expressed as a combination of interior values and boundary data; they may not be introduced additional unknowns. Science there are no nodes outside the boundary, these approximations must be based on one-sided differences or approximations.

Usually, convective fluxes are prescribed at the inflow boundary. Convective fluxes are zero at impermeable walls and symmetry planes, and are usually assumed to be independent of the coordinate normal to an outflow boundary. The specification of these conditions varies according to the type of boundary and the dependent variables under consideration. The treatment and implementation of boundary conditions are given in great details in Versteeg and Malalasekera (1995). Here, a brief description is provided.

3.3.1 Inlet boundary

At inlet boundary, all quantities have to be prescribed. The values of variables are assigned for each grid node at the entry plane. These values must be known from measurements, from the problem specifications or from estimates of conditions prevailing in the inflow stream. The mean flow properties at the inlet are usually known, while some or all turbulence properties have to be estimated. In the absence of Reynolds stresses at the inlet, the turbulent kinetic energy is estimated as:

\[ k_{in} = \frac{3}{2} (I_i U_{in})^2 \]  

(3.6)
where, $U_\text{in}$ is bulk velocity, and $I_t$ is the turbulence intensity with values ranging form 1%-10%. The scalar dissipation rate $\varepsilon$ on the other hand has to be estimated and is usually estimated from length scale as:

$$\varepsilon_{in} = \frac{C^{3/4} k_{in}^{3/2}}{l_m}$$  \hspace{1cm} (3.7)$$

where $l_m$ is the turbulent mixing length. For a pipe flow, $l_m = 0.07 \times \text{Radius}$

### 3.3.2 Plane or axis of symmetry

In many flows there are one or more symmetry planes or axis of symmetry. The symmetric solution can be obtained by solving the problem in part of the solution domain using the symmetry conditions. At a symmetry plane the normal components of the convective and diffusive fluxes are zero i.e. there is no transport across a plane of symmetry. The normal velocity component is zero, but its normal gradient is not; thus the normal stress $\tau_{nn}$ is non-zero. This boundary condition can be implemented by assigning the appropriate coefficients $a_i$ to zero.

### 3.3.3 Outlet boundary

At the outlet we usually know little about the flow. The outlet boundaries should be placed as far downstream of the region of interest as possible so that the flow is fully developed and is in the direction out of the solution domain. The usual approach is to use upwind approximation at the out-flow surface and then to apply zero gradient condition at the boundary. This is sufficient for all variables except for the normal velocity. Overall continuity imposes a constraint and the outlet velocity should be such that the overall mass conservation is preserved over the complete solution domain. One way to ensure this is to calculate the mass flux going out of the domain $M_{out}$ by first applying zero gradient. The velocity at the outlet is then scaled by the multiplying factor $M_{in}/M_{out}$, where $M_{in}$ is the inlet mass flow rate.
3.3.4 Solid walls

At an impermeable solid wall the no slip condition follows from the fact that viscous fluid sticks to solid boundary and no slip condition is the appropriate boundary condition at a solid wall. Since there is no flow through the wall, convective fluxes of all quantities are zero. However, the turbulent flow over a solid surface is characterised by the presence of a very thin viscous sublayer adjacent to the wall along which rapid variations of the mean velocity and turbulent properties exist. The number of grid points required to resolve the viscous sublayer is prohibitively large. The log-law of the wall is used to overcome the mesh resolution problem. The implementation of the log-law condition has been done in many different ways (Versteeg and Malalasekera 1995; Jones 1994). Table 3.2 shows the optimal near wall relationships according to Versteeg and Malalasekera (1995). These relationships are used in conjunction with the universal log-law of the wall:

\[ u^+ = \frac{1}{\kappa} \ln(Ey^+) \]  

(3.8)

Some of the notations of the near wall relationships of Table 3.2 are given in Fig. 3.3, which shows near-boundary CV and a u-cell near the solid wall. The relationships of Table 3.2 are implemented in the discretised equations through the source terms \( S_C \) and \( S_p \).

**Table 3.2**: Near wall relationship for the \( k - \varepsilon \) model

<table>
<thead>
<tr>
<th>Equation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Momentum equation tangential to the wall:</td>
<td></td>
</tr>
<tr>
<td>wall shear stress ( \tau_w = \rho C_{1/4}^{1/4} k_{p}^{1/2} \frac{u_p}{u^+} )</td>
<td></td>
</tr>
<tr>
<td>wall force ( F_s = -\tau_w A_{cell} = -(\rho C_{1/4}^{1/4} k_{p}^{1/2} \frac{u_p}{u^+}) A_{cell} )</td>
<td></td>
</tr>
<tr>
<td>Momentum equation normal to the wall:</td>
<td></td>
</tr>
<tr>
<td>normal velocity = 0.0</td>
<td></td>
</tr>
<tr>
<td>Turbulence kinetic energy:</td>
<td></td>
</tr>
<tr>
<td>generation of turbulent kinetic energy, ( G = (\tau_w u_p - \rho C_{3/4}^{3/4} k_{p}^{3/2} u^+) \frac{\Delta V}{\Delta y_p} )</td>
<td></td>
</tr>
<tr>
<td>Dissipation rate equation:</td>
<td></td>
</tr>
<tr>
<td>net nodal value, ( \varepsilon_p = C_{3/4}^{3/4} k_{p}^{3/2} / (\kappa \Delta y_p) )</td>
<td></td>
</tr>
</tbody>
</table>
3.4 Solution algorithm

The equations for mass conservation, momentum and the coupled scalar variables are solved by using the SIMPLE algorithm (Patankar 1980). The description of the SIMPLE algorithm can be found in many texts (Patankar 1980; Versteeg and Malalasekera 1995; Ferziger and Peric 1996) and is not repeated here. The combustion models are incorporated into the algorithm as follows:

1. Guess the initial conditions for all variables.
2. Update the boundary conditions.
3. Solve the momentum equations.
4. Solve the pressure correction equations.
5. Correct the pressure and velocities.
6. Solve the k equation.
7. Solve the ε equation.
8. Solve the other scalar equations (for combustion modelling).
9. Calculate the density.
10. Use the new values as the initial conditions and repeat steps 2 to 8 until convergence is reached.
11. Calculate the temperature and mass fraction of species after convergence is reached.
The set of algebraic equations are solved by repeated sweeps of a line by line application of the well known tri-diagonal matrix algorithm (TDMA) (Patankar 1980). The equations are solved along constant \( x_i \) and in doing so the variables located along adjacent lines are kept constant. The coefficient \( a_i \) and source term, \( S_C \) in Eq.(3.2) are evaluated using values of the variables at the start of the TDMA sweep and are not changed during the TDMA solution. The solution is regarded as converged when the residual source term scaled against a representative inflow flux is small (typically \( 10^{-5} \)) for all variables.

The combustion models are incorporated in the steps 8, 9 and 11 of the algorithm and are described below.

### 3.4.1 Flame sheet/Equilibrium model

The incorporation of the flame sheet and equilibrium models in the general solution algorithm is similar. The relationship, \( \phi = \phi(Z) \), calculated either by the flame sheet model or by the equilibrium chemistry model, is tabulated in a file. The data file is used as an input to the CFD code. As explained earlier, the beta pdf used to incorporate the influence of the turbulent fluctuations is given by Jones (1994):

\[
P(Z) = \frac{Z^{a-1}(1-Z)^{b-1}}{\int_0^1 Z^{a-1}(1-Z)^{b-1} \, dZ} = \frac{Z^{a-1}(1-Z)^{b-1}}{\Gamma(a)\Gamma(b)\Gamma(a+b)}
\]

where the values of the exponents \( a \) and \( b \) are given by:

\[
a = \tilde{Z} \left[ \frac{\tilde{Z}(1-\tilde{Z})}{\tilde{Z}^2} - 1 \right]
\]

\[
b = (1-\tilde{Z}) \frac{a}{\tilde{Z}}
\]

and where, the mean, \( \tilde{Z} \), and the variance, \( \tilde{Z}^2 \), of the mixture fraction are obtained from the solution of their respective transport equations.
Chapter 3. Numerical Implementation

Because $P(Z)$ is a density-weighted pdf, mean values are given by:

$$
\bar{\phi} = \frac{1}{\Gamma(a+b)} \int_0^1 \phi(Z)P(Z)dZ = \frac{\Gamma(a+b)}{\Gamma(a)\Gamma(b)} \int_0^1 \phi(Z)Z^{a-1}(1-Z)^{b-1}dZ
$$

(3.12)

$$
\bar{\phi} = \bar{\rho} \int_0^1 \frac{\phi(Z)}{\rho(Z)} P(Z)dZ
$$

(3.13)

where

$$
\bar{\rho} = \left[ \int_0^1 \frac{P(Z)}{\rho(Z)} dZ \right]^{-1}
$$

(3.14)

and the deviations of $\phi$ from the mean values are estimated from:

$$
\left[ \frac{\bar{\rho}}{\int_0^1 \frac{(\phi(Z) - \bar{\phi})^2}{\rho(Z)} P(Z)dZ} \right]^{1/2}
$$

(3.15)

The flame sheet/equilibrium model enters the general solution algorithm in steps 8, 9 and 11 as follows:

**Step 8:**

8a: Solve the transport equation for mean mixture fraction, Eq. (2.60)

8b: Solve the transport equation for mean mixture fraction variance, Eq.(2.61).

**Step 9:**

9: Calculate the density from Eq. (3.14).

**Step 11:**

11: Calculate the temperature, mass fraction of species and other scalar variables and their deviations from Eq. (3.13) and (3.15).
The integration in Eqs.(3.12), (3.13) and (3.14) is evaluated by the Romberg's method with the midpoint approximation (Press et al. 1993). The $\phi(Z)$ at the intermediate point of the data file required for the integration is obtained from interpolation. The end points ($Z=0,1$) of the integration become singular in the numerical calculation procedure when the parameter $a$ or $b$ is less than unity. This singularity is eliminated analytically according to the method suggested by Bray et al. (1994) and Chen et al. (1994). According to them, the integration in Eq.(3.12) is approximated by:

$$\int_{0}^{1} \phi(Z)Z^{a-1}(1-Z)^{b-1}dZ \approx \frac{\xi^a}{a} \phi(0)$$

$$+ \int_{\xi}^{1} \phi(Z)Z^{a-1}(1-Z)^{b-1}dZ + \frac{\xi^b}{b} \phi(1)$$

(3.16)

where, $\xi$ represents a very small number (say $10^{-30}$).

Another numerical difficulty is that the computed value of $a$ and $b$ in Eqs.(3.10) and (3.11) may approach magnitudes of several hundred thousands in the iteration process (Chen et al. 1994). This problem in turn leads to overflow in the calculation of $P(Z)$. According to the characteristic of the beta function, the $P(Z)$ will be close to a delta function when either value of exponent $a$ and $b$ is adequately large (Fig. 3.4). In the present calculation, when the value of $a$ or $b$ is very large (say, 500), the $P(Z)$ is approximated by a delta function, $P(Z) = \delta(Z - \bar{Z})$. The mean scalar variable is then given by:

$$\bar{\phi} = \int_{0}^{1} \phi(Z)P(Z)dZ = \int_{0}^{1} \phi(Z)\delta(Z - \bar{Z})dZ = \phi(\bar{Z})$$

(3.17)
3.4.2 Laminar flamelet model

The laminar flamelet model has been described in section 2.6.2. According to this model, the scalar variables depend on two parameters, the mixture fraction and the scalar dissipation rate, so that $\bar{\phi} = \phi(Z; \chi)$. The mean value of the scalar variable is given by:

$$\bar{\phi} = \int_0^\infty \int_0^1 [\phi(Z; \chi)P(Z)P(\chi)] dZd\chi$$

(3.18)

The double integration in Eq.(3.18) is simplified by using a limited number of flamelet libraries each represented by the scalar dissipation rate $\chi_l$. The integration range in $\chi$ is divided into $L$ subranges $[\chi_{l-1/2}, \chi_{l+1/2}]$, with $l=1,2,...,L$ (in particular $\chi_{1/2} = 0$), such that $\chi_l$ is a representative value for the corresponding interval. The integration is evaluated by means of the approximation (Lentini 1994):
The integration of \( \int_0^1 \phi(Z; \chi) \) is similar to that explained in the previous section. The evaluation of \( \int_{\chi_{l-1/2}}^{\chi_{l+1/2}} P(\chi) d\chi \) is explained here. The integration requires the specification of the pdf shape in \( \chi \) space. As mentioned earlier, the pdf \( P(\chi) \) is assumed to be log-normal:

\[
P(\chi) = \frac{1}{\chi \sigma \sqrt{2\pi}} \exp\left( -\frac{1}{2\sigma^2} (\ln \chi - \mu)^2 \right)
\]  

(3.20)

where the parameters \( \mu \) and \( \sigma \) are related to the mean value of \( \chi \) by:

\[
\tilde{\chi} = \exp(\mu + \frac{1}{2} \sigma^2)
\]  

(3.21)

The integration \( \int_{\chi_{l-1/2}}^{\chi_{l+1/2}} P(\chi) d\chi \) is thus simplified in the form:

\[
\int_{\chi_{l-1/2}}^{\chi_{l+1/2}} P(\chi) d\chi = \frac{1}{\sqrt{\pi}} \int_{\theta_{l-1/2}}^{\theta_{l+1/2}} e^{-\theta^2} d\theta
\]  

(3.22)

\[
= \frac{1}{2} [\text{erf}(\theta_{l+1/2}) - \text{erf}(\theta_{l-1/2})]
\]

where \( \text{erf} \) denotes the error function. Finally, the mean value of density and scalar variable given by Eq. (3.17) is rewritten as:

\[
\bar{\rho} \left[ \sum_{l=1}^{L} \frac{1}{2} [\text{erf}(\theta_{l+1/2}) - \text{erf}(\theta_{l-1/2})] \int_{0}^{1} \frac{P(Z)}{\rho(Z, \chi_l)} dZ \right]^{-1}
\]  

(3.23)

\[
\bar{\phi} = \sum_{l=1}^{L} \frac{1}{2} [\text{erf}(\theta_{l+1/2}) - \text{erf}(\theta_{l-1/2})] \int_{0}^{1} \phi(Z, \chi_l) P(Z) dZ
\]  

(3.24)
\[ \overline{\phi} = \overline{\rho} \left[ \sum_{l=1}^{L} \frac{1}{2} [\text{erf}(\theta_{l+1/2}) - \text{erf}(\theta_{l-1/2})] \int_{0}^{1} \frac{\phi(Z, \chi_l)}{\rho(Z, \chi_l)} P(Z) dZ \right] \]  \hspace{1cm} (3.25) \\

and the deviations of \( \phi \) from the mean values are estimated from:

\[ \left[ \overline{\rho} \sum_{l=1}^{L} \frac{1}{2} [\text{erf}(\theta_{l+1/2}) - \text{erf}(\theta_{l-1/2})] \int_{0}^{1} \frac{(\phi(Z, \chi_l) - \overline{\phi})^2}{\rho(Z, \chi_l)} P(Z) dZ \right]^{1/2} \hspace{1cm} (3.26) \]

where arguments of the error function are given explicitly in terms of the computed quantities as follows:

\[ \ln \left( \frac{\chi_{l+1/2}}{\chi} \right) + \frac{1}{2} \sigma^2 \hspace{1cm} (3.27) \]

and the scalar dissipation rate is given by:

\[ \tilde{\chi} = C_{\chi} \frac{\overline{\varepsilon}}{k} \overline{Z^2} \hspace{1cm} (3.28) \]

The flamelet library is represented by the inert mixing state for \( \chi > \chi_q \) indicating an extinguished flamelet. In order to accurately account for the contribution of the extinguished flamelet, the integration range is divided into subranges in such a way that \( \chi_{L-1/2} = \chi_q \) and \( \chi_{L+1/2} = \infty \). Further details of the integration method can be found in (Lentini 1994).

The laminar flamelet model is incorporated into the general algorithm in steps 8, 9 and 11 as follows:

**Step8:**
- 8a: Solve the transport equation for mean mixture fraction, Eq. (2.60)
- 8b: Solve the transport equation for mixture fraction variance, Eq. (2.61)

**Step9:**
- 9a: Calculate the scalar dissipation rate from Eq. (3.28).
9b: Calculate the arguments of the error function from Eq.(3.27).

9c: Calculate the density from Eq. (3.23).

Step 11:
11: Calculate the temperature, mass fraction of species and other scalar variables and their deviations from Eq. (3.25) and (3.26).

3.4.3 Eddy break-up model

The eddy break-up model is an empirical model for expressing the mean chemical reaction rate in the transport equations of the mean mass fractions. The EBU model is described in section (2.6.3). Here, the numerical implementation of the EBU model in the CFD code is described. The EBU model enters in steps 8, 9 and 11 as follows:

Step 8:
8a: Solve the transport equation for the mean enthalpy, Eq.(2.92).

8b: Solve the transport equations for the mean mass fraction of the species, Eqs.(2.89), Eqs.(2.90) and Eqs.(2.91).

Step 9:
9: Calculate the density from the equation of state, Eq.(2.94).

Step 11:
11: Calculate the temperature from Eq.(2.93) and other scalar variables.

With the EBU model, an ignition procedure is required as initially the product of combustion is absent throughout the flow field. The ignition is achieved by assigning a small amount of mass fraction of product near the fuel jet entry.

Although the eddy-break-up model has proved rather successful in predicting overall combustion rates, its total neglect of the chemical kinetics has disbarred it from simulating secondary but important kinetically-controlled processes, such as NOX production in flames.
Flamelet Library

The laminar flamelet model requires as an input of library of flamelet profiles. The accuracy of the model in many ways depends on the accuracy of the flamelet library. Experimental data can be used as the flamelet library as in Liew et al. (1984). However, more detailed information can be obtained by generating the flamelet profiles from the numerical solution of laminar flames. As explained earlier, there are two different approaches available for calculating the flamelet profiles. The first method is based on the numerical solution of a counterflow diffusion flame (Peters 1984; Rogg et al. 1986; Drake et al. 1988; Lentini and Puri 1995). The second method is the solution of the flamelet equations based on the universal co-ordinate transformation (Peters 1986; Bray and Peters 1994; Pitsch and Peters 1998; Lentini 1994).

In the present study, flamelet profiles are calculated using both methods for CO/H$_2$/N$_2$ flame and for CH$_4$/H$_2$ flame only the counterflow diffusion flame method has been employed.

4.1 Computation of flamelet library

The mathematical formulations for generating flamelet library are given in section 2.6.2. The flamelet libraries used in the simulation of turbulent non-premixed flames of the present study consist of 7 flamelets. Liew et al. (1984), Sanders and Lamers (1994) and Gran et al. (1994) have used 2 flamelets in their calculations, Sanders and Gökalp (1997) have used 5 flamelets, Lentini (1994) have used 7 flamelets and Drake et al. (1988) have used 17 flamelets. The use of higher numbers of flamelets will certainly improve accuracy, but it will also increase the computational time considerably. The laminar flame and flamelet code RUN-1DL (Rogg 1995) is used to compute the
flamelet library. In the numerical procedure, the equations governing the laminar flamelet problem are discretised using the finite difference technique. The resulting equations are solved on adaptive grid system, in which additional grids are added at locations where the profiles of the variables to be solved have large gradients or curvatures. The adaptive gridding strategy in the code is designed to produce a mesh that resolves the thin flame front with a reasonable number of grid points.

The flamelet equations are solved with detailed reaction mechanism of Warnatz (Peters 1993). The reaction mechanism is given in appendix A. The reaction mechanism involves 12 species (CO, O2, CO2, H2O, H2O2, HO2, H, OH, O, H2, HCO, N2) for CO/H2/N2 fuels and 17 species (CH4, O2, H2O, CO2, CO, H2, H, OH, O, HO2, CH3, HCO, CH2O, H2O2, CH, CH2, N2) for CH4/H2 fuel.

The flamelet profiles calculated by the counterflow diffusion flame method are on the physical space, which should be converted to the mixture fraction space to use in the laminar flamelet modelling. The mixture fraction can be defined in terms of atomic masses of elements present in the system as:

\[ Z = \frac{Z_m - Z_{m,o}}{Z_{m,F} - Z_{m,o}} \]  \hspace{1cm} (4.1)

where \( Z_m \) is the mass fraction of element \( m \) and subscripts \( O \) and \( F \) refer to air and fuel streams respectively. Different researchers have used different elements for the definition of mixture fraction. Lentini and Puri (1995) have used N-element and Sanders and Gökalp (1997) have used H-element for the definition of mixture fraction. However, the mixture fractions calculated from the C, H, O and N element mass fractions generally differ by some amount because of the differences in the molecular transport of different species (Drake and Blint 1988). Bilger (1988) proposed an overall mixture fraction which is a linear combination of the elemental mixture fractions. Bilger's formula is used in the present study:
where subscripts $O$ and $F$ refer to air and fuel streams respectively. $W_\beta$ is the atomic weight of element $\beta$. This definition yields the correct stoichiometric mixture fraction, even in the presence of the differential diffusion effect. Bilger's formula has been widely accepted for the definition of mixture fraction in the laminar flamelet modelling (Pitsch and Peters 1998) as well as analysing the experimental data (Masri et al. 1996; Dally et al. 1998a) in the past few years.

The scalar dissipation rate is another parameter to characterise the flamelets. For the counterflow geometry, the scalar dissipation rate is approximated as a function of mixture fraction, assuming constant density and diffusivity by (Bray and Peters 1993): 

$$\chi = \frac{a}{\pi} \exp\left\{ -2erfc^{-1}(Z)^2 \right\}$$

(4.3)

where $a$ is the strain rate and $erfc^{-1}$ is inverse of the complementary error function.

Clearly, the scalar dissipation rate varies along the counterflow flame, but it is convenient to have a single value of the scalar dissipation rate to represent each flamelet. The scalar dissipation rate at the stoichiometric location is used in the present study to represent each flamelet. In some studies, the value of the scalar dissipation rate at the maximum temperature has been used (Liew et al. 1984; Lentini 1994). Choosing the value of the scalar dissipation rate at the maximum temperature is not expected to provide significant differences in turbulent combustion simulation results.

### 4.2 Flamelet library for CO/$H_2$/N$_2$ flame

The fuel considered here consists of 27.5% CO, 32.3% H$_2$ and 40.2% N$_2$ by volume. The counterflow diffusion flame on Tsuji burner is simulated by keeping the normalised
fuel velocity fixed at 6.4, while varying the strain rate at the air stream side. Dixon et al. (1984) have shown that the normalised fuel velocity (Eq. 2.72) only affects the location of the stagnation point and the flame relative to the cylinder without disturbing the flame structure. The flamelet profiles for temperature and mass fractions of CO, H₂, H₂O, CO₂ and OH are shown. The flamelet profiles are generated with full molecular transport of the species and for different strain rate values. The corresponding scalar dissipation rates are indicated in the figures 4.1-4.9. The peak temperatures at the scalar dissipation rates of $\chi_{st} = 0.026$/s and $\chi_{st} = 0.02576$/s are higher than the equilibrium temperature of 2161 K. The peak temperatures in excess of equilibrium temperature at lower scalar dissipation rate are caused by the differential diffusion effect. The preferential diffusion of H₂ leads to the enrichment of H₂ at the flame front relative to the initial fuel mixture and subsequently results in higher temperature. Only for higher scalar dissipation rate does the flamelet temperature fall below the equilibrium temperature. The effect of stretching of the flamelets is clearly evident from these figures. The influence of the scalar dissipation rate on the temperature is high. The peak temperature falls from 2415 K at $\chi_{st} = 0.026$/s to 1375 K at $\chi_{st} = 1262.24$/s. The effect of scalar dissipation rate on the H₂ and CO mass fractions is very small and its effect is only observed near the stoichiometric mixture fraction. The H₂O mass fraction profiles are moderately influenced by the scalar dissipation rate. The CO₂ mass fraction on the other hand is considerably influenced by the scalar dissipation rate. The peak CO₂ is reduced by almost 50% at the extinction limit and also the location of the peak shifts towards the fuel rich zone. With the increase of scalar dissipation rate, the OH level first increases and then starts to decrease as the scalar dissipation rate approaches the extinction limit. The peak values of OH for $\chi_{st} = 0.026$/s and $\chi_{st} = 0.1262.24$/s are similar, but at $\chi_{st} = 0.1262.24$/s, the width of the OH profile is much higher.

In order to investigate the effect of differential diffusion, the flamelet profiles are generated by setting $Le=1.0$ for all the species. The flamelet profiles are obtained for the strain rate values of 0.1, 10.0, 100.0, 2000.0 and 4000.0 /s with corresponding scalar dissipation rate of 0.026, 2.576, 25.76, 128.80, 515.20 and 1030.4 /s. The extinction limit is reduced for the unity Lewis number case as compared to the differential diffusion case. The flamelet profiles
for the unity Lewis number are shown in Figs. 4.4-4.6. The peak temperature at the lowest scalar dissipation rate is 2151 K, which is 10 K below the equilibrium temperature of 2161 K. With the increase of scalar dissipation rate, the location of the peak temperature shifts towards the fuel rich side. The peak temperature at the extinction condition is almost equal to that with the full molecular transport. The level of H₂O profiles decreases by about 30%, while the level of CO₂ increases by about 60% as compared to the flamelets with full molecular transport. The OH flamelet profiles show slight reduction of the peak values when compared to those of the flamelet with the differential diffusion effect.

The flamelet profiles generated by the flamelet equations in the conserved scalar space are shown in Figures 4.7-4.9. In this method, the scalar dissipation rate has to be specified as an input data. Calculations are performed for the values of χₘ of 0.48, 3.66, 33.27, 155.41, 600.46 and 1436.0 /s. The last value corresponds to the extinction limit χₑ. The peak temperature at the lowest scalar dissipation rate is 2023 K, which is less than the equilibrium temperature of 2161 K. The flamelet profiles for species concentrations are similar to those obtained by the counterflow diffusion flame with unity Lewis number.

Figure 4.1: Flamelet profiles of temperature and mass fraction of CO for different scalar dissipation rate obtained from counterflow diffusion flame method with differential diffusion effects. — — χ = 0.026; — — — χ = 2.576; — — — — χ = 25.76; — — — — χ = 128.80; — — — — — χ = 515.20; — — — — — — χ = 1262.24 /s
Figure 4.2: Flamelet profiles of mass fraction of H$_2$ and H$_2$O for different scalar dissipation rate obtained from counter flow diffusion flame method with differential diffusion effects. —— $\chi = 0.026$; —— $\chi = 2.576$; ——— $\chi = 25.76$; ——— $\chi = 128.80$; ——— $\chi = 515.20$; ———— $\chi = 1262.24$ /s

Figure 4.3: Flamelet profiles for mass fraction of CO$_2$ and OH for different scalar dissipation rate obtained from counter flow diffusion flame method with differential diffusion effects. —— $\chi = 0.026$; —— $\chi = 2.576$; ——— $\chi = 25.76$; ——— $\chi = 128.80$; ——— $\chi = 515.20$; ———— $\chi = 1262.24$ /s
Figure 4.4: Flamelet profiles of temperature and mass fraction of CO for different scalar dissipation rate obtained from counter flow diffusion flame method with unity Lewis number. —— $\chi = 0.026$; — $\chi = 2.576$; —— $\chi = 25.76$; —— $\chi = 128.80$; — $\chi = 515.20$; —— $\chi = 1030.40$ /s

Figure 4.5: Flamelet profiles of mass fraction of $\text{H}_2$ and $\text{H}_2\text{O}$ for different scalar dissipation rate obtained from counter flow diffusion flame method with unity Lewis number. —— $\chi = 0.026$; — $\chi = 2.576$; —— $\chi = 25.76$; —— $\chi = 128.80$; — $\chi = 515.20$; —— $\chi = 1030.40$ /s
Figure 4.6: Flamelet profiles of mass fraction of CO₂ and OH for different scalar dissipation rate obtained from counter flow diffusion flame method with unity Lewis number. —— \( \chi = 0.026 \); —— \( \chi = 2.576 \); ——— \( \chi = 25.76 \); ——— \( \chi = 128.80 \); ——— \( \chi = 515.20 \); ——— \( \chi = 1030.40 \) /s

Figure 4.7: Flamelet profiles of temperature and mass fraction of CO for different scalar dissipation rate obtained from flamelet equations in mixture fraction space. —— \( \chi = 0.048 \); —— \( \chi = 3.66 \); ——— \( \chi = 33.27 \); ——— \( \chi = 155.41 \); ——— \( \chi = 600.46 \); ——— \( \chi = 1436.34 \) /s
Figure 4.8: Flamelet profiles of mass fraction of H₂ and H₂O for different scalar dissipation rate obtained from flamelet equations on mixture fraction space. —— χ = 0.048; --- χ = 3.66; ——— χ = 33.27; ———— χ = 155.41; ———— χ = 600.46; ———— χ = 1436.34 /s

Figure 4.9: Flamelet profiles of mass fraction of CO₂ and OH for different scalar dissipation rate obtained from flamelet equations on mixture fraction space. —— χ = 0.048; --- χ = 3.66; ——— χ = 33.27; ———— χ = 155.41; ———— χ = 600.46; ———— χ = 1436.34 /s
4.3 Flamelet library for CH₄/H₂ flame

The CH₄/H₂ fuel considered here consists of 50% CH₄ and 50% H₂ by volume. The flamelet profiles are generated for the strain rates of 3.0, 20.0, 100.0, 500.0, 1800.0, 3600.0 /s (extinction value) with the full molecular transport and 3.0, 20.0, 100.0, 500.0, 1000.0, 1800.0 /s (extinction value) with the equal mass diffusivities flamelets. The extinction limit is halved when equal mass diffusivities are assumed for this flame. Figures 4.10-4.14 and 4.15-4.19 respectively show the flamelet profiles with and without the differential diffusion effects. Unlike CO/H₂/N₂ fuel, CH₄/H₂ fuel is less influenced by the scalar dissipation rate. The peak temperature is reduced by about 600 K for the differential diffusion flamelets and by about 400 K for the unity Lewis number flamelets. The peak temperature at the extinction condition is about 1800 K with and without the differential diffusion effect. The effect of the differential diffusion is observed in varying degrees for temperature and mass fractions of H₂O, OH and CO. The mass fractions of H₂ and OH are slightly reduced and the mass fraction of CO is slightly increased in the unity Lewis number flamelets as compared to the differential diffusion flamelets. The CO₂ levels are almost doubled for the unity Lewis number case. The peak mass fraction of CO₂ for this fuel is located at the stoichiometric mixture fraction, Z=0.05, both with and without differential diffusion effect. The straight line patterned gradual decay towards zero value at Z=1.0 in the mass fraction of CO₂ observed between Z=0.1 and Z=1.0.
Figure 4.10: Flamelet profiles of temperature for different scalar dissipation rate obtained from counter flow diffusion flame method with differential diffusion effects. $\chi = 0.064; \cdots \chi = 0.428; \cdots \cdots \chi = 2.139; \cdots \cdots \chi = 10.695; \cdots \cdots \chi = 38.505; \cdots \cdots \cdots \chi = 77.01 /s$

Figure 4.11: Flamelet profiles of mass fraction of H$_2$O for different scalar dissipation rate obtained from counter flow diffusion flame method with differential diffusion effects. $\chi = 0.064; \cdots \chi = 0.428; \cdots \cdots \chi = 2.139; \cdots \cdots \chi = 10.695; \cdots \cdots \chi = 38.505; \cdots \cdots \cdots \chi = 77.01 /s$
Figure 4.12: Flamelet profiles of mass fraction of CO$_2$ for different scalar dissipation rate obtained from counter flow diffusion flame method with differential diffusion effects. $\chi = 0.064; \chi = 0.428; \chi = 2.139; \chi = 10.695; \chi = 38.505; \chi = 77.01 /s$

Figure 4.13: Flamelet profiles of mass fraction of OH for different scalar dissipation rate obtained from counter flow diffusion flame method with differential diffusion effects. $\chi = 0.064; \chi = 0.428; \chi = 2.139; \chi = 10.695; \chi = 38.505; \chi = 77.01 /s$
Figure 4.14: Flamelet profiles of mass fraction of CO for different scalar dissipation rate obtained from counter flow diffusion flame method with differential diffusion effects. —— \( \chi = 0.064 \); ——- \( \chi = 0.428 \); ———- \( \chi = 2.139 \); ———— \( \chi = 10.695 \); ————- \( \chi = 38.505 \); ————--- \( \chi = 77.01 \) /s

Figure 4.15: Flamelet profiles of temperature for different scalar dissipation rate obtained from counter flow diffusion flame method with unity Lewis number. —— \( \chi = 0.064 \); ——— \( \chi = 0.428 \); ———- \( \chi = 2.139 \); ———— \( \chi = 10.695 \); ————- \( \chi = 38.505 \); ————--- \( \chi = 77.01 \) /s
Figure 4.16: Flamelet profiles of mass fraction of H$_2$O for different scalar dissipation rate obtained from counter flow diffusion flame method with unity Lewis number. --- $\chi = 0.064$; -- $\chi = 0.428$; --- $\chi = 2.139$; --- $\chi = 10.695$; ---- $\chi = 38.505$; ----- $\chi = 77.01$ /s

Figure 4.17: Flamelet profiles of mass fraction of CO$_2$ for different scalar dissipation rate obtained from counter flow diffusion flame method with unity Lewis number. --- $\chi = 0.064$; -- $\chi = 0.428$; --- $\chi = 2.139$; --- $\chi = 10.695$; ---- $\chi = 38.505$; ----- $\chi = 77.01$ /s
Figure 4.18: Flamelet profiles of mass fraction of OH for different scalar dissipation rate obtained from counter flow diffusion flame method with unity Lewis number. \(-\chi = 0.064; \cdots \chi = 0.428; \cdots \cdots \chi = 2.139; \cdots \cdots \chi = 10.695; \cdots \cdots \chi = 38.505; \cdots \cdots \chi = 77.01 \text{ /s}\)

Figure 4.19: Flamelet profiles of mass fraction of CO for different scalar dissipation rate obtained from counter flow diffusion flame method with unity Lewis number. \(-\chi = 0.064; \cdots \chi = 0.428; \cdots \cdots \chi = 2.139; \cdots \cdots \chi = 10.695; \cdots \cdots \chi = 38.505; \cdots \cdots \chi = 77.01 \text{ /s}\)
Modelling of Transport and Chemical Kinetics of H$_2$/N$_2$ Jet Flames

In this chapter, the prediction of fuel species concentrations and temperatures of H$_2$/N$_2$ jet flames are presented. Results obtained with different combustion models are compared to one another and to the available experimental data. The different cases of H$_2$/N$_2$ jet flames are investigated for different inlet velocities and Reynolds numbers to study the effect of turbulence-chemistry interaction. These flames are well suited for the evaluation of combustion models as practical combustion situations. This study presents further insights into the structure of the H$_2$/N$_2$ jet diffusion flames. Combustion is modelled using the infinitely fast chemistry and finite rate chemistry assumptions. The evaluation of combustion models for the different cases of the flames are presented. Overall, predictions of major and minor species, and flame temperatures, are in reasonable agreement with data. The presented results demonstrate that the flamelet models predictions based on the differential diffusion as well as with unity Lewis number are, in general, both qualitatively and quantitatively superior to those of other models. Overall predictions of major and minor species and flame temperatures, are in reasonable agreement with experimental data. Results do, however, tend to underpredict or overpredict mixture fractions, temperatures and species mass fractions in some cases. The effects of differential diffusion on the flamelet model predictions are analysed. The sensitivity of the laminar flamelet models to different non-equilibrium parameters are presented next. Finally, some important aspects of the application of combustion models are discussed and conclusions are drawn. This work extends and validates the use of standard models of turbulence to symmetric jets and flames. These results are in an improved understanding of the structure of such flows and flames.
5.1 Jet Burner and Flame Definitions

Three flame cases namely HN1, HN2 and HN3 have been simulated and the results have been shown in figures 5.2–5.47. Here the symbol HN stands to denote the Hydrogen and Nitrogen flame for convenience and 1,2,3 are indications of the different cases of flames. The HN1 flame consists of 75% H₂ + 25% N₂ by volume and has a jet velocity of 94.0 m/s with Re=10300. The three H₂/N₂ jet flames under investigation in this chapter are with different initial conditions and experimental data for these flames can be accessed from Sandia TNF webpage. The flames are unpiloted and issue from 4.0-mm diameter nozzle for HN1 and 8.0-mm diameter nozzle for HN2 and HN3 flames, centred at the exit of a vertical wind tunnel of internal diameter 140mm, 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, with a summary of other initial conditions being given in table 5.1.

Table 5.1: Initial conditions for the investigated H₂/N₂ jet diffusion flames

<table>
<thead>
<tr>
<th>Flame</th>
<th>H₂: N₂ by volume</th>
<th>Re</th>
<th>Exit velocity m/s</th>
</tr>
</thead>
<tbody>
<tr>
<td>HN1</td>
<td>75:25</td>
<td>10300</td>
<td>94.0</td>
</tr>
<tr>
<td>HN2</td>
<td>75:25</td>
<td>9300</td>
<td>42.3</td>
</tr>
<tr>
<td>HN3</td>
<td>50:50</td>
<td>6200</td>
<td>21.7</td>
</tr>
</tbody>
</table>

The Reynolds numbers are calculated from the bulk velocity at the burners exit where the temperature is 300 K. These flames are well suited for the evaluation of combustion models as a model problem and practical combustion situations.

5.2 Uncertainty and error levels in Measurements

To evaluate any numerical model predictions quantitatively against the experimental data it is very important to know the error associated with the measurements. The combination of spontaneous Raman scattering and Rayleigh scattering are used in most
of the flame experiments to measure the major species concentrations (N2, O2, CO, H2, CO2, H2O) and temperature. Linear LIF are used to measure the concentrations of OH and NO. Fluorescence signals were corrected on a shot-to-shot basis for variations in the Boltzmann fraction and collisional quenching rate, based on measured temperature and major species concentrations in the probe volume. Collisional quenching cross sections for OH and NO were reported in the work of Paul et al. (1994). The error associated with the diffusion flame were resulted from many factors such as shot noise, electronic noise, error associated with optical set up, spatial resolution error, the cross talk between the Raman signals, the fluorescence interference from soot precursors and other molecules and the interpolation of the Raman calibration factors.

Representative values of precision of the single-point measurements given in Dally et al. (1998a) for two typical samples of a CH4/H2 flame are reproduced in table 5.2. The errors associated with the interference and the spatial resolutions are not given, as these errors are difficult to quantify (Dally et al. 1998a). The fluorescence interference affects only selected species. The fluorescence interference from soot precursors is very low for these flames. Hydrocarbon flames are most affected by the interference and CO line suffers the highest interference levels. The error associated with fluoresce interference is generally less than 10%. The spatial resolution error estimates are also given in Dally et al. (1998a). According to them, the maximum resolution error is 9% near nozzle exit. At all locations where the error is less than 4%. The maximum error associated with the NO measurements was reported to be approximately 10% (Dally et al. 1998a).
Table 5.2: Sample estimates of error associated with measurements of species concentrations sample diffusion flames

<table>
<thead>
<tr>
<th>Sample</th>
<th>Temperature (K)</th>
<th>Species</th>
<th>Mass Fraction (%)</th>
<th>Error (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lean</td>
<td>1900</td>
<td>O₂</td>
<td>4.0</td>
<td>10.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>N₂</td>
<td>75.0</td>
<td>0.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CO₂</td>
<td>8.0</td>
<td>4.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CO</td>
<td>2.0</td>
<td>9.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>H₂</td>
<td>0.5</td>
<td>12.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>H₂O</td>
<td>11.0</td>
<td>5.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>OH</td>
<td>0.3</td>
<td>3.8</td>
</tr>
<tr>
<td>Rich</td>
<td>1400</td>
<td>CH₄</td>
<td>18.0</td>
<td>2.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td>N₂</td>
<td>57.0</td>
<td>1.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CO₂</td>
<td>5.5</td>
<td>5.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CO</td>
<td>5.5</td>
<td>8.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td>H₂</td>
<td>2.5</td>
<td>4.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>H₂O</td>
<td>12.0</td>
<td>4.0</td>
</tr>
</tbody>
</table>

The maximum error associated with the NO measurements was reported to be approximately 10% (Dally et al. 1998a).

5.3 Numerical Simulations and Initial conditions

Computational methods for the present flames were performed using modified TEACHT based CFD code together with the flame simulation code RUN-1DL, developed by Rogg (1995). RUN-1DL code used to generate flamelet libraries for the flames. The flame sheet, equilibrium and laminar flamelet models are incorporated in the CFD code. Some results were obtained using the commercially available CFD software FLUENT. The choice was based on familiarity with the package and on the fact that it is widely used in industrial and academic purposes. The code has undergone extensive testing and its widespread use in industry makes it particularly useful for this type of study. The model allows flow field and combustion calculations to be performed in complex 3D geometries. All calculations were made assuming steady mean flow. Favre averaging is used throughout. The ‘standard k-ε’ model of turbulence employed to incorporate the turbulent effects.
The constants used in these models are explained in the previous chapter along with the PDF weighted equations for the mass fraction and temperature employed in the reacting case. The conservation equations are solved using a finite-volume method, which uses a nonstaggered grid with all variables stored at control volume centres. The performance of the turbulence models was initially checked for parabolic flows with a round jet. A modification to one of the constants in the turbulence model proposed by Morse (1977) and Pope (1978) is discussed and validated in this study (see section 5.4), the modified model was then applied unchanged to the elliptic flows of jet burner.

The experimental data can only be fully exploited for the evaluation of the numerical models if reliable and accurate boundary conditions are available for use in the simulation. The specification of the boundary conditions at symmetry axis, solid wall and outlet plane is well defined as explained in section 3.3. The initial profiles of the mean velocity and the Reynolds-stress tensor components are fitted by the use of experimental data near the nozzle, in correspondence to round jet flow profiles. The mean mixture fraction is set to a Heavyside unit step function and the scalar variance is zero at the whole inlet. The Dirichlet boundary conditions are prescribed on the outer boundaries, symmetric boundary conditions are applied on the centerline.

### 5.4 Jet flow and turbulent constants

It has been established that the \( k-\varepsilon \) and RS models overpredict the decay rate and the spreading rate of a round jet flow according to Pope (1978) and Rodi (1984, 1986). The means of overcoming this problem in the \( k-\varepsilon \) and RS models are well known: the constants \( C_\mu \) (the constant in the turbulent viscosity), \( C_{e1} \) and \( C_{e2} \) (the constants for generation/destruction of the \( \varepsilon \) term) are made functions of the velocity decay rate and the jet width Pope (1978). Such modifications to the transport dissipation equation include those of McGuirk and Rodi (1979) who suggested the following modification of the \( C_{e1} \) constant in the dissipation equations:

\[
C_{e1} = 1.14 - 5.31 \frac{y_{1/2}}{U_{cl}} \frac{dU_{cl}}{dx}
\]

Where the retardation parameter, \( (y_{1/2}/U_{cl})dU_{cl}/dx \), is used, where \( U_{cl} \) is the centerline velocity and \( y_{1/2} \) is the distance from the axis where the velocity is half the centerline.
value. McGuirk and Rodi (1979) noted that for self-similar round jets the retardation parameter takes a value of $-0.087$, while for a self-similar plane jet the parameter is $-0.055$. These values lead to the $C_{e1}$ constant having a value of 1.44 in the plane jet and 1.6 in the round jet.

Similar to the above formulation, Morse (1977) proposed the following modification to the $C_{e1}$ constant as follows:

$$C_{e1} = 1.4 - 3.4 \left( \frac{k}{\varepsilon} \frac{\partial U_{cl}}{\partial x} \right)_{cl}^3$$

This modification does not use the width of the jet and relies only on the decay rate of the centerline velocity. It is noteworthy that all the modifications suggested for $C_{e1}$ lead to a value of 1.6 for self-similar round jets.

Another modification to the $C_{e2}$ constant was suggested by Launder et al. (1972). This modification calculates the $C_{e2}$ constant as:

$$C_{e2} = 1.92 - 0.667 \left( \frac{v_{1/2}}{2U_{cl}} \left( \frac{dU_{cl}}{dx} - \frac{dU_{el}}{dx} \right) \right)^{0.2}$$

The above modifications were designed for parabolic flows and are not immediately applicable to elliptic flows.

Pope (1978) has questioned the generality of these modifications and suggested that the stretching of turbulent vortex tubes by the mean flow has a significant influence on the process of scale reduction. He argues that in flow regions where the mean vorticity is being stretched, so also is the turbulent vorticity, leading to greater scale reduction, greater dissipation, less kinetic energy and hence to a lower effective viscosity. According to this argument, the effective viscosity and the spreading rate are lower in a round jet than in a plane jet. He also proposed the addition of an extra term $\psi$ to the dissipation equation, which accounts for the non-dimensional stretching of the vortices. This term would also have an arbitrary multiplier which is set to $C_{e3} = 0.79$ for a simple jet. The dissipation equation with this modification then becomes

$$\frac{D\varepsilon}{Dt} = \frac{\partial}{\partial x_i} \left( \frac{\mu_{eff}}{\sigma} \frac{\partial \varepsilon}{\partial x_i} \right) + \frac{\varepsilon^2}{k} \left( C_{e1} \frac{P}{\varepsilon} - C_{e2} + C_{e3} \psi \right)$$
where, $\psi$ is the vortex stretching invariant for axisymmetric flows without swirl, is

$$
\psi = \frac{1}{4} \left( \frac{k}{\epsilon} \right)^3 \left( \frac{\partial U}{\partial r} - \frac{\partial V}{\partial x} \right)^2 \frac{V}{r}
$$

The modifications suggested by Morse (1977) and Pope (1978) were implemented in the $k$-$\epsilon$ model and tested against the experimental data of Barlow et al (1998). Both required the use of very low underrelaxation factors for $k$ and $\epsilon$ (0.007) as compared with 0.2-0.6 when a constant value of $C_{\epsilon 1} = 1.60$ is used. However using a very large number of iterations (~ 10000) good convergence was obtained. The mean centerline velocities and their RMS fluctuations plotted against the axial distance above the exit plane are shown in figure 5.1.

![Figure 5.1: Comparison of the modified $k$-$\epsilon$ turbulence models predictions of centerline velocity and its rms with experimental data for the CO/H$_2$/N$_2$ jet flame. Standard $k$-$\epsilon$ model and $k$-$\epsilon$ model with $C_{\epsilon 1} = 1.60$ (lines); measurements (circles) [Barlow et al (1998)].](image)

Comparisons of computational results were made with measured data in this figure. Results are shown for the standard $k$-$\epsilon$ model ($C_{\epsilon 1} = 1.44$) as well as for the modified
ones ($C_{e1} = 1.60$). Significant improvement is obtained in the results from computations using the new constant. This test confirms the validity of the modification of $C_{e1}$ as applied to the jet flame problems.

5.5 Evaluation of combustion models

Comparisons and evaluation of results obtained from different combustion models have been made in this section. The combustion models are evaluated by comparing radial profiles of mean mixture fraction, variance of mixture fraction, temperature and concentrations of major and minor species at different axial locations. The combustion models assessed are the flame sheet, equilibrium and flamelet models. In flamelet model unity Lewis number and the effects of differential diffusion are considered.

5.5.1 HN1 Flame

The HN1 flame as shown in table 5.1 consists of 75% $\text{H}_2 + 25\% \text{N}_2$ by volume and has a jet velocity of 94.0 m/s with $Re=10300$. Calculated mixture fraction, temperatures and concentrations are given in Favre averaged values. The mean and r.m.s. values of mixture fraction, mass fractions and temperature along the axis cross-section profiles at $(x/D=5), (x/D=10), (x/D=20), (x/D=40), (x/D=60),$ and $(x/D=80)$ are presented.

Based on the results of the simulations, a number of general remarks can be made on the characteristic of the interactions between the turbulent flow field and the chemistry. From the predicted results of the mass fraction of $\text{H}_2, \text{N}_2, \text{O}_2$ and to a certain extent $\text{H}_2\text{O}$, it can be argued that the relatively simple equilibrium, the flame-sheet models are able to produce good predictions. However, that cannot be argued for the temperature prediction. From the physical and chemical point of view, temperature is a very important quantity. In many practical applications, it is essential to obtain a correct temperature field. The overall agreement between prediction and measurement is satisfactory. However, the flamelet model performs better predictions of temperature profiles. This is mainly due to the better prediction of the mixture fraction field.

Figure 5.2 shows radial profiles of the mixture fraction at six axial locations. It is essential that the flow fields are adequately represented in the simulations, because the
mixture fraction statistics completely determine the thermo-chemistry of the flame. It is even more important for the flames with low stoichiometric mixture fraction \( (Z_{st} = 0.08) \) like \( \text{H}_2/\text{N}_2 \) jet flame. A small error in mixture fraction field for this fuel will lead to a large error in temperature, other species calculations and in the location of the flame front. At \( x/D=5 \), the core of the outer region contains fuel rich mixture and the stoichiometric mixture fraction exist at the edge of the flame front. The flamelet models produce better predictions than flame sheet and equilibrium models for the mixture fraction profiles. Though the radial mixture fraction is reasonably well predicted by all the combustion models in the fuel rich zone until \( x/D=40 \), a small amount of underprediction is observed in the midstream of the flame by flamelet model whereas the improvements in flame sheet and equilibrium models predictions are prominent in mid to the downstream of the flame.

Radial profiles of the mixture fraction variance at six axial locations are shown in Fig.5.3. The overall predictions of all the combustion models are fairly good. At \( X/D=5 \), the maximum variance overpredicted slightly and little bit down position along radial distance by models than that of experimental position. Further down level, the mixture fraction variance is overpredicted by all the combustion models until it nullified. At \( X/D=10 \), good predictions are observed by models upto the peak variance and flame sheet and equilibrium models show better predictions than that of flamelet models. At the locations \( X/D=20 \) and 40, all the models predict good but flamelet models shown slightly better results for the later. At \( X/D=60 \) and 80, the flamelet model with differential diffusion predicted reasonably the best for all radial position than all the other models. For the axial location \( X/D=20 \) to 80 the mixture fraction variances show a gradual decay inside the core zone of the flame.

Radial profiles of the mass fractions of \( \text{H}_2 \), \( \text{N}_2 \) and \( \text{O}_2 \) at six axial locations are shown in Figs.5.4-5.6. These profiles are similar to the mixture fraction profiles. The overall flamelet models predictions of \( \text{H}_2 \) are better with the exception of results at the locations \( X/D=05 \) and 20 where flamesheet and equilibrium models respectively have shown better predictions. All the combustion models overpredicted the \( \text{N}_2 \) profile at \( X/D=05 \), whereas the predictions at all other locations are reasonable with better flamelet models results. The slight underpredictions in \( \text{O}_2 \) profiles have shown by all the models except equilibrium model. The profiles of \( \text{O}_2 \) are better predicted by the
flamesheet model at some locations. The flamelet model with the differential diffusion effect predicts the O$_2$ profiles better than the flamelet model with unity Lewis number.

Figure 5.7 shows the temperature profiles at different axial locations. At x/D = 05, the measured mean flame tip given by the maximum temperature is located around the radial distance 3mm whereas the calculated flame tip position is slightly away from the centreline than that of experiment. This corresponds to the slower decay of mixture fraction in the calculations. The temperatures are well predicted by all the models except the equilibrium model, and the predicted peak temperatures are within 300K for different models at all the locations except at x/D=20. At this location, the peak temperature prediction differences are in the range of 300K-600K by different combustion models. The differences in temperature predictions in the simulations and that the lower measured temperature as compared to calculations is due to the averaging effects as a result of the intermittency in the flame at these locations and the underprediction is not due to the shortcomings in the simulations. Further from the nozzle, radial temperatures obtained from flamelet models improve over other predictions, however that the temperature profiles decay too rapidly over the radius, with this effect being accentuated with downstream distance.

Figure 5.8 shows radial profiles of mass fraction of H$_2$O. The flame sheet and equilibrium model predicts well upto the peak value of H$_2$O for x/D= 5, 10 and 20 whereas the flamelet models agree well with the data further down level along radial distances. For x/D= 40 to 80 flamelet models have the domination of agreement level of predictions with experiment. Other models also show the reasonable results.

Radial profiles of the RMS fluctuations of the mass fractions of H$_2$, N$_2$ and O$_2$ at six axial locations are shown in Figs.5.9-5.11. The flame sheet and equilibrium models failed to predict the maxima of the fluctuations of H$_2$ and overpredicted the results unrealistically. The flamelet models with differential diffusion and with unity Lewis number show the overall better predictions. In the predictions of the rms of N$_2$ and O$_2$, the peak levels fluctuations are observed little bit down levels along radial directions except at x/D=5 of N$_2$ prediction. The flamesheet and equilibrium models have shown higher level of fluctuations and the flamelet models shown lower levels. But in most of
the cases the flamesheet and equilibrium models predicted well near the leading edges and flamelet models have shown better results for down levels along radial directions.

Figure 5.12 shows the radial profiles of the RMS fluctuations temperature profiles at different axial locations. The peak levels of temperature fluctuations are predicted slightly away along radial directions by all the models. For x/D = 05 to 40 flamesheet and equilibrium models perform better in predictions of temperature fluctuations from the centreline to the maximum fluctuations and the flamelet models do so from the peak value towards down level until the fluctuations nullified. At x/D = 60 and 80 flamelet models perform better all over the region whereas all the models failed to predict the peak fluctuations correctly at all locations.

Radial profiles of the RMS fluctuations of the mass fractions of H$_2$O at six axial locations are shown in Figure 5.13. At X/D=05 there are two maximum fluctuations. The first predicted maximum value obtained slightly closer to the jet centreline than that of the data but the second one obtained at the same position as experimental data. The predictions at this location are performed better by flamelet model with differential diffusion and also by the flame sheet model. At X/D=10 there are two maximum fluctuations as well. The first predicted peak value obtained at the right position as the data but the second one obtained away from the jet centreline than that of the experimental data. The first peak value predictions at this location are performed better by flamelet models but none of the models could predict the second peak value properly. For further away from the centreline all the models have shown reasonable agreement with the data for both the axial locations. At X/D=20 the peak levels of rms of H$_2$O mass fractions are predicted slightly away from the jet centreline than experimental data and none of the models could predict the result exactly but for further away all the model predictions agreed with the experiment. At x/D = 40 flame sheet and equilibrium models performed better until it reached to the peak value and then flamelet models have shown better results. At x/D = 60 and 80 though the flamelet models predictions are in better agreement with data all the models have shown reasonably good results.
The overall agreement between predictions and measurements are satisfactory. However, the flamelet model performs better. This is mainly due to the better prediction of the mixture fraction field. The effects of turbulence-chemistry interaction are analysed. All calculated values have the same order of magnitude and shape as the measured data. The correspondence is very well and the reasonable levels of agreements are achieved in the mixture fraction, mass fractions and temperature predictions. This gives the evidence, that coupling of chemical models with turbulence are well suited for the HN1 flame.
Figure 5.2: Radial profiles of mean mixture fraction in the HN1 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu) i.e. Le=1.0.
Figure 5.3: Radial profiles of mixture fraction variance in the HN1 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.4: Radial profiles of mass fraction of H₂ in the HN1 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.5: Radial profiles of mass fraction of N$_2$ in the HN1 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMd), flamelet with unity Lewis number (FLMu)
Figure 5.6: Radial profiles of mass fraction of $O_2$ in the HN1 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.7: Radial profiles of temperatures in the HN1 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.8: Radial profiles of mass fraction of H$_2$O in the HN1 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.9: RMS fluctuations of H$_2$ in the HN1 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)

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X/D=05, X/D=10, X/D=20, X/D=40, X/D=60, X/D=80
Figure 5.10: RMS fluctuations of N$_2$ in the HN1 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.11: RMS fluctuations of $O_2$ in the HN1 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.12: RMS fluctuations of temperature in the HN1 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.13: RMS fluctuations of H$_2$O in the HN1 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
5.5.2 HN2 Flame

The HN2 flame consists of 75% H₂ + 25% N₂ by volume and has a jet velocity of 42.3 m/s with Re=9300 as described in table 5.1. The comparison between experimental data and predicted results are shown in Figs.5.14 – 5.25 for this flame. The mean and r.m.s. values of the profiles along the axis cross-section at (x/D=5), (x/D=10), (x/D=20), (x/D=40), (x/D=60), and (x/D=80) are presented.

Figure 5.14 shows the mean mixture fraction profiles at six axial locations. The overall agreements between the predictions and the measurements are reasonably good by all the combustion models. However, there is slight overpredictions in the regions 2< r< 10 mm at x/D=0.5, 2 < r< 14 mm at x/D=10, 2 < r< 20 mm at x/D=20, 5 < r< 30 mm at x/D=40 by all the models. At x/D=60 flamesheet and equilibrium models have overpredicted the mixture fraction for almost all the radial positions. The slight differences are also observed between the predicted results of different combustion models near the centre line at all the six locations.

Radial profiles of the mixture fraction variance at six axial locations are shown in Fig.5.15. As in the HN1 flame the overall predictions of all the combustion models are fairly good for all the axial locations. At X/D=5, the maximum variance found around the radial locations 3.5 ≤ r≤ 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 further away form the centreline along radial direction. At the locations X/D=20 and 40, good predictions are observed with the exception of slightly overprediction of the peak values. At X/D=60 and 80, all the models have predicted well and shown the gradual decay of fluctuation in mixture fraction along the radial direction. The overall better predictions have been observed in the results obtained by the flamelet model with differential diffusion.

Radial profiles of the mass fractions of H₂, N₂ and O₂ at six axial locations are shown in Figs.5.16-5.18. These profiles are similar to the mixture fraction profiles. The overall predictions of H₂ by the models are good with the exception of results at the locations
X/D=40 and 80 where flamesheet and equilibrium models respectively have shown slight overpredictions. At X/D=80 experimental data shows no level of H2 but the models predictions have shown very little amount of existence of the Hydrogen. The flamelet model with differential diffusion predicted the N2 profile well at X/D=05 with small amount of underprediction. The flamesheet and equilibrium models overpredicted the N2 profile except the flamesheet model provide a good prediction for r> 9mm. At X/D=10, the flamelet models whereas the predictions at all other locations are reasonable with better flamelet models provide good results with slight overprediction in the region r<10 and underprediction for r>10. Results from the other models are fare. Similar results have been obtained for X/D=20. At X/D=40 all the models overpredicted the N2 profiles but the flamelet model with differential diffusion predicted well only for 9<r<53mm. At X/D=60 flamelet models provided good predictions with slight overprediction near the centreline upto r = 40mm and other models have shown higher overprediction. At the location X/D=80 flamelet model with differential diffusion predicted well for the radial location between 0.0 and 100mm and for the further down level the flamelet model with unity Lewis number and equilibrium model provide good results. The flamelet models provide very good predictions in mass fraction of O2 profiles at all axial locations. Other models have also shown the fairly good results with slight overpredictions. The profiles of O2 are better predicted by flamelet model with the differential diffusion at the locations X/D= 05, 40, 60 and 80 but at the other locations better predictions are observed in the results obtained by the flamelet model with unity Lewis number.

Figure 5.19 shows the temperature profiles at six axial locations. At x/D = 05, the temperature is reasonably well predicted by all the combustion models. However, the flamelet models undepredicted the temperature in the fuel rich side of the flame. The peak temperature also predicted very near to the measured location. At this axial location the maximum measured temperature is about 2250K which is 400K higher than that of HN1 flame at the same location, this is because of the higher ratio of the reaction time scale to turbulent time scale due to lower jet exit velocity in the HN2 flame. At x/D = 10, the maximum temperature underpredicted at the location away from centreline by all the models than that of experiment. The predicted temperature profiles have shown rapid decay from the maximum position similar to the experimental data.
The peak temperature is overpredicted by 400K and little bit away from the centreline at \(x/D=20\) by the equilibrium model but underpredicted between 100 to 200K. At \(x/D=40, 60\) and 80, the agreements between the measurements and the predictions are very good with slight overprediction by the flamesheet and equilibrium models in the fuel lean zones. In the laminar flamelet model, local extinction is predicted when the scalar dissipation rate reaches a quenching limit and the blow-off of the flame occurs when the proportion of burning flamelets decreases below a certain limit. However, the flamelet model in the present study shows that this simple treatment is not adequate to predict the local extinction and subsequent lower temperatures in the jet flames.

Figure 5.20 shows radial profiles of mass fraction of H\(_2\)O at different axial locations. As it was in the HN\(_1\) flame the flame sheet and equilibrium models predicted well up to the peak value of H\(_2\)O for \(x/D= 5\) and 10 whereas the flamelet models have shown better prediction for the decaying region. For \(x/D= 20\) and 40 flamelet models provide good predictions from centreline towards the peak value and all the models overpredicted the results in the decaying region of the mass fraction of H\(_2\)O. At \(x/D= 60\) and 80 flamelet models predicted the H\(_2\)O profile very well and other models have also shown the reasonable results with slight overpredictions.

Radial profiles of the RMS fluctuations of the mass fractions of H\(_2\), N\(_2\) and O\(_2\) at six axial locations are shown in Figs.5.21-5.23. The flame sheet and equilibrium models have overpredicted the fluctuations of H\(_2\) mass fractions but the flamelet models predicted better. At \(x/D=80\) all the models predicted three maximum values whereas measurements identified only two. In the predictions of the rms of N\(_2\), the peak levels fluctuations are overpredicted by the flame sheet and equilibrium models observed little bit away from the centreline than that of measurements. The flamelet models predicted well with slight underprediction but at the same radial location as the other models predictions. The maximum values of the fluctuations of O\(_2\) mass fractions at X/D=05, 10, 20 and 40 predicted closer to the centreline than experimental positions and models predictions are not good at the first three axial locations but at the last three axial locations overall predictions are comparatively better.
Figure 5.24 shows the radial profiles of the RMS fluctuations temperature profiles at different axial locations. There are two maximum fluctuations in temperatures at \( x/D = 0.5 \) and 10 but none of the models could predict those well. At \( x/D = 20 \) and 40 the maximum fluctuation levels predicted very close to the measurements but flamelet models underpredicted slightly and overpredicted at \( x/D = 20 \) by flamesheet and equilibrium models. The flamesheet and equilibrium models predicted the peak level well at \( X/D = 40 \) but the flamelet models have shown overall better predictions at this location and other two axial locations as well.

Radial profiles of the RMS fluctuations of the mass fractions of H\(_2\)O at six axial locations are shown in Figure 5.25. At \( X/D=0.5, 10 \) and 20 all the models predicted well from centreline towards the peak level and overpredicted for further away. The peak levels predicted slightly away from the jet centreline than that of the data. Similarly, at \( X/D= 40 \) the peak levels of rms of H\(_2\)O mass fractions are predicted slightly away from the jet centreline than experimental data. The flamelet models predicted well but other models have shown overpredictions for most of the radial locations. At \( X/D= 60 \) and 80 the flamesheet and equilibrium models overpredicted the rms values of H\(_2\)O mass fractions highly near the centreline and the error decreases gradually with the increasing radial distance. The flamelet models provided good predictions for all over the radial locations and reproduced the pattern very well. The overall agreement between predictions and measurements are satisfactory with the better flamelet models performances also the couplings of chemical models with turbulence are well suited for the HN\(_2\) flame.
Figure 5.14: Radial profiles of mean mixture fraction in the HN2 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with Le=1.0 (FLMu).
Figure 5.15: Radial profiles of mixture fraction variance in the HN2 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.16: Radial profiles of mass fraction of $H_2$ in the $HN_2$ flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.17: Radial profiles of mass fraction of N\textsubscript{2} in the HN\textsubscript{2} flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.18: Radial profiles of mass fraction of O$_2$ in the HN$_2$ flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.19: Radial profiles of temperatures in the HN2 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.20: Radial profiles of mass fraction of H$_2$O in the HN$_2$ flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.21: RMS fluctuations of $H_2$ in the HN2 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.22: RMS fluctuations of N\textsubscript{2} in the H\textsubscript{2}/N\textsubscript{2} flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.23: RMS fluctuations of $O_2$ in the HN2 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.24: RMS fluctuations of temperature in the HN2 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.25: RMS fluctuations of H$_2$O in the HN2 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
5.5.3 HN3 Flame

This flame consists of 50% H\textsubscript{2} + 50% N\textsubscript{2} by volume and has a jet velocity of 21.7 m/s with Re=6200 as described in table 5.1. The comparison between experimental data and combustion models predicted results are shown in Figs.5.26–5.37 for this flame. The mean and r.m.s. values of the profiles along the axis cross-section at (x/D=2.5), (x/D=10), (x/D=20), (x/D=30), (x/D=50), and (x/D=70) are presented.

Figure 5.26 shows the mean mixture fraction profiles for different axial locations. The location of the flame front is situated where Z\textsubscript{st} = 0.08 and is well predicted by the flamelet models for all the axial locations. At x/D=2.5 the agreements between the predictions and the measurements are good for flamelet and flamesheet models near the centreline upto r=4.5mm but overpredictions are observed in the regions 5 < r < 7.5 mm by all the combustion models. At x/D=10, there are overpredictions by all the combustion models between r = 2 and r = 12 mm. The flamelet models predicted well at the locations x/D=20, 30, 50 and 70. The other models have also shown overpredicted results at x/D=20, 30, 50 but reasonably good results observed at x/D=70.

Radial profiles of the mixture fraction variance at six axial locations are shown in Fig.5.27. All the combustion models failed to predict maximum level of fluctuations as well as the other values properly for the axial locations x/D=2.5 and 10. At X/D=20 and 30, combustion models underpredicted the mixture fraction variance from the centreline towards the peak value and then good predictions are observed further away from the peak level with slight overprediction. At the locations X/D=50 and 70, better agreement are observed near the centreline and slight overpredictions found for further down region.

Radial profiles of the mean mass fractions of H\textsubscript{2}, N\textsubscript{2} and O\textsubscript{2} at six axial locations are shown in Figs.5.28-5.30. The H\textsubscript{2} mass fractions well predicted near the centreline by the models at the locations X/D=2.5, 10, 20 and 30. The flamelet models predicted well at X/D=20 and slight overprediction observed in the core zones at the other three locations. At X/D=50 and 70 very close predictions provided by the models with slight overpredictions. The flamelet and flamesheet models predicted the N\textsubscript{2} profile well at X/D=10 only and at all the other locations predicted results obtained from all the
models are in the state of overpredictions. At X/D=20 and 30 results from the flamelet model with unity Lewis number are very near to the measurements. The flamelet models provide good predictions at all locations in mass fraction of O$_2$ profiles except slight overpredictions in the core zone around the flamefront region at X/D= 2.5, 10, 20 and 30. The other models overpredicted the results at all axial locations.

Figure 5.31 shows the temperature profiles at six axial locations. None of the models could predict the maximum temperature properly at x/D = 2.5, 10 and 20. Most of the models tend to underpredict the temperature profiles from centreline towards the maximum and overpredict further away along the radial distance. Equilibrium model predicted the peak value correctly at x/D = 2.5 but it shifted the predicted peak location slightly away from the centreline like all other models. At this axial location the performance of flamesheet model is very good upto the measured peak position but it overpredicted the peak value itself including all other values further away along the radial distance. At x/D = 10 flamesheet model predicted well in the near axis region and flamelet model predicted well in the far axis region. At x/D = 20 flamesheet model overpredicted thoroughly, equilibrium model also overpredicted with slight exception in the near axis region and flamelet model predicted well in the far axis region but under predicted in the near axis region. At x/D = 30, the flamelet models predicted well and other models have shown reasonable results with slight overpredictions. At x/D=50 and 70 all the models overpredicted the results but flamelet models provided very close prediction to the measurements.

Figure 5.32 shows radial profiles of mass fraction of H$_2$O at different axial locations. All the models failed to predict the location of the maximum level of the H$_2$O mass fraction, at x/D= 2.5 and 20 and observed slightly away from the centreline as that of experiments. All the models tend to underpredict the H$_2$O profiles from centreline towards the maximum and overpredict further away along the radial distances at the locations x/D = 2.5, 10 and 20. For x/D= 30, 50 and 70 flamelet models provide very good predictions in H$_2$O mass fraction profiles and other models have shown reasonable results with slight overpredictions.

RMS fluctuations of the mass fractions of H$_2$, N$_2$ and O$_2$ at six axial locations are shown in Figs.5.33-5.35. At X/D=2.5, all the models have predicted the fluctuations of H$_2$ mass fractions well between the region of positive gradients upto the extreme point and overpredictions observed along the decaying region. All the models failed to
predict the maximum fluctuations of \( H_2 \) and their radial positions for \( x/D=10, 20 \) and 30 also overpredictions found along the decaying region. At \( x/D=50 \) and 70 no \( H_2 \) fluctuations are there in the experimental data but very little amount of \( H_2 \) fluctuations with wavy effect are observed in the models predictions. In the predictions of the rms of \( N_2 \), at \( x/D=2.5, 10 \) and 20 the peak levels fluctuations are overpredicted by the flame sheet and equilibrium models observed little bit away from the centreline than that of measurements. The flamelet models predicted better with slight underpredictions. At \( x/D=40, 50 \) and 70 the flamelet models predicted better with wavy effects and other models provide overpredicted results. The maximum values of the fluctuations of \( O_2 \) mass fractions at \( x/D=2.5, 10, 20 \) and 30 predicted away from the centreline than experimental positions. The flamesheet and equilibrium models predictions have shown higher maximum values at the first three axial locations but flamelet models predicted better with slight underpredictions. For the last two axial locations overall predictions are tend to overestimate with flamelet models better predictions.

Figure 5.36 shows the radial profiles of the RMS fluctuations of temperature profiles at different axial locations. There are two maximum fluctuations in temperatures at \( x/D = 2.5 \) which are predicted well by equilibrium model at forward shifted positions along the radial distance. No other models could predict those well. The flamesheet and equilibrium models predicted well near the centreline upto the first maximum fluctuation and flamelet models underpredicted these values but well predicted further down next to the second maximum and therefore, it is difficult to draw a conclusion about the predictive capabilities of the combustion models based on the predicted results at this location. At \( x/D = 10 \) and 20 none of the models could predict the maximum fluctuation levels well and tend to overpredict the results along the fuel lean region. At \( x/D = 30 \) flamelet model with unity Lewis number predicted the maximum fluctuation well at slightly forward shifted position along the radial distance. Both the flamelet models predictions are fairly good in the near and far axis region. At \( x/D = 50 \) and 70 all the models overpredicted the fluctuations of temperature but flamelet model with unity Lewis number provide comparatively closer results to the measurements.

Radial profiles of the RMS fluctuations of the mass fractions of \( H_2O \) at six axial locations are shown in Figure 5.37. At \( X/D=2.5 \) the flamesheet and equilibrium models predicted well from centreline towards the peak level and overpredicted for further away but flamelet models predicted well in the decaying. At \( X/D=10 \) all the models
overpredicted the H$_2$O fluctuations in the decaying region region and none of them could predict the maximum level properly but flamelet model with unity Lewis number provided overall better results. At X/D= 20 and 30, the flamelet models provided good predictions but failed to predict the peak level of fluctuation at x/D= 20 and suffered from under prediction. At X/D= 50 and 70, all the models overpredicted the fluctuations of H$_2$O mass fractions but they reproduced the trend of the fluctuations of the H$_2$O mass fractions well. The overall agreement between predictions and measurements are satisfactory with the better flamelet models performances.
Figure 5.26: Radial profiles of mean mixture fraction in the HN3 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with Le=1.0 (FLMu).
Figure 5.27: Radial profiles of mixture fraction variance in the HN3 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.28: Radial profiles of mass fraction of H$_2$ in the HN3 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.29: Radial profiles of mass fraction of N$_2$ in the HN3 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.30: Radial profiles of mass fraction of $O_2$ in the HN3 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.31: Radial profiles of temperatures in the HN3 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.32: Radial profiles of mass fraction of H$_2$O in the HN3 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.33: RMS fluctuations of H$_2$ in the HN3 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.34: RMS fluctuations of N₂ in the HN₃ flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.35: RMS fluctuations of O\textsubscript{2} in the HN3 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.36: RMS fluctuations of temperature in the HN3 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
Figure 5.37: RMS fluctuations of H$_2$O in the HN3 flame. Experimental (EXP), flame sheet model (FLS), equilibrium (EQL), flamelet with differential diffusion (FLMdd), flamelet with unity Lewis number (FLMu)
5.6 Effects of non-equilibrium parameters

The parameters currently used in the flamelet modelling to describe non-equilibrium effects in are: (i) the scalar dissipation rate and (ii) the strain rate. The results obtained from the flamelet model with unity Lewis number by using different non-equilibrium parameters are presented for the HN2 flame.

The mean strain rate is calculated from $\bar{s} = \bar{\varepsilon}/\bar{k}$ according to Bray et al. (1994). However, Sanders and Lamers (1994) and Sanders Gökalp (1995) defined the mean strain rate by $\bar{s} = 6.4\bar{\varepsilon}/\bar{k}$. In the HN2 flame, this definition would give the mean strain rates much above the extinction value in the zone close to the nozzle exit, which is unrealistic. The fluctuations of the strain rate are incorporated by a quasi Gaussian distribution (Sanders and Lamers 1994). A modified scalar dissipation rate defined by Bray et al. (1994) is also compared here. Bray et al. (1994) argued that the flamelets are subjected to the fluctuations of the order of square of the flamelet thickness, $(\Delta Z)^2$. The effective mean scalar dissipation rate acting on the flamelet is then $\chi = 2(\Delta Z)^2 \sigma^2/k$; where the flame thickness is $\Delta Z = 2Z_{st}$.

The results obtained by employing different non-equilibrium parameters are compared in Figs. 5.38 and 5.39 for temperature and mass fraction of H$_2$O respectively. The temperatures are severely underpredicted near the exit plane zone by the strain rate formulation indicating localised extinction. The strain rate is very high at the flame front, whereas the value of the scalar dissipation rate is low at same location. This is because the scalar dissipation rate depends on both the strain rate and the mixture fraction variance and the value of mixture fraction variance is low at the outer edge. These two formulations predict completely opposite phenomenon; the scalar dissipation formulation predicts a flame closer to equilibrium, but the strain rate formulation predicts a locally extinguished flame. The prediction by the modified scalar dissipation rate is better than that of the strain rate, but worse than the predictions by the standard scalar dissipation rate. At $x/D=0.5$, the prediction errors are very high and errors decrease gradually along the axial downstream levels. The predicted temperatures are almost similar at $x/D=60$ and $x/D=80$ for all the non-equilibrium formulations. The predictions of mass fraction of H$_2$O by different non-equilibrium parameter are similar to those of the temperature.
Figure 5.38: Effects of non-equilibrium parameters on the prediction of temperatures in the HN2 flame. • measurements; — standard scalar dissipation rate; --- modified scalar dissipation rate; ---- by strain rate.
Figure 5.39: Effects of non-equilibrium parameters on the prediction of H$_2$O mass fractions in the HN$_2$ flame. o measurements; —— standard scalar dissipation rate; —— modified scalar dissipation rate; ——- by strain rate.
5.7 Effects of discretisation schemes

The results presented in this chapter are obtained by employing the hybrid scheme for discretisation of the convection term in the governing equations. The error associated with the discretisation scheme is assessed by employing two more schemes, power law and total variation diminishing (TVD) scheme with Van Leer and UMIST limiters. The grid sizes are adapted to get the best possible output in all the discretisation schemes.

The results obtained by using different schemes and grid size for the HN3 flame are compared in figures 5.40 – 5.42. In the calculations of the presented figures, the combustion is accounted for by the laminar flamelet model with unity Lewis number.

The variations among the predicted results of mixture fraction, mixture fraction variance and temperature are small. The maximum difference is observed in the peak temperature at x/D=0.9. The TVD scheme with the UMIST limiter produces a 3.1% decrease of the peak temperature and the hybrid scheme produces very small variations in the peak temperatures with other schemes. These variations are well below the accuracy of the measurements. Therefore, it can be concluded that the spatial discretisation errors are small enough to allow the underlying combustion models to be evaluated by any one of the aforementioned discretisation schemes. The hybrid scheme has been used to evaluate the combustion models in this chapter as a consequence.
Figure 5.40: Effects of discretisation schemes on the prediction of mixture fractions in the HN3 flame at different axial positions along with the contour. Measurements; --- hybrid scheme; ---- power law; ---- TVD scheme with Van Leer limiter; ---- TVD scheme with UMIST limiter.
Figure 5.41: Effects of discretisation schemes on mixture fraction variance in the HN3 flame at different axial position. o measurements; ——— hybrid scheme; ——— power law; ——— TVD scheme with Van Leer limiter; ———— TVD scheme with UMIST limiter.
Figure 5.42: Effects of discretisation schemes on temperatures in the HN3 flame at different axial position. • measurements; —— hybrid scheme; — — power law; ———- TVD scheme with Van Leer limiter; ——- TVD scheme with UMIST limiter.
5.8 Discussion

The following text presents discussion of some results of H$_2$/N$_2$ jet diffusion flames of different exit velocity and compositions. Figure 5.43 shows the contours of mixture fractions of HN1, HN2 and HN3 flames. The momentum flux of the fuel jet has a strong influence on the flow structure of the combustion and flame zones. The effects of differences in exit velocities and fuel compositions on the mixture fraction contours and flame fronts are prominent clearly visible in the figures of contours. The increase of jet momentum also results in a longer flame as observed from the stoichiometric contours. It is apparent that the mixture fraction field in the upper and intermediate jet velocity flows show similar contours with the different height of flame tips and location of stoichiometric mixture fraction contours. This is because they have the same fuel composition and the only difference is the exit velocity. Similar phenomenon can be observed in the contours of temperatures and H$_2$O mass fractions.

The influence of the momentum flux of the fuel jet on the burning pattern of the flames are also clearly shown in the contour plots of temperatures as shown in Figure 5.44 (a) HN1 flame, (b) HN2 flame, (c) temperature contours of HN3 flame with half side grid overlay. However these grids had been refined accordingly and adapted to obtain the best possible outcome from the simulation. The peak temperatures near the exit plane for all the flames are very close to the flame axes and are for small regions of radial distances which are not seems to be appeared in the contours. But in the further downstream region, different contours are very clear to identify the temperature zones. Whereas the temperature levels are clearly shown in the line plots against the data. The HN1 flame shows peak temperatures at the edge of the reaction zones and outside of it. The temperature contours for the HN2 and HN3 flames are however similar to the HN1 flame. All the contour plots of temperature also show that the reaction between x/D $\approx$ 2.0 and x/D $\approx$ 60 do not reach chemical equilibrium condition.

The contours of mass fractions of H$_2$O for HN1, HN2 and HN3 flames are shown in figure 5.45. The rate of productions of H$_2$O are very small in the near axis and near exit plane region and it increase gradually until the x/D = 30 or around x/D = 25. The effects
of exit velocities and fuel compositions on the mass fraction contours are also prominent in the figures.

Figure 5.46 shows the predictions of the temperatures for HN1, HN2 and HN3 flames at two axial locations x/D = 10 and x/D = 20. Plotted against the experimental scatter data are the flamelet predictions with constant mass diffusivities and unity Lewis number and with different values of strain rates. For the sake of clarity, it should be noted the scatter data observable in the locale of the mixture fraction space and are shown for two axial locations only. Significant good predictions of temperatures are observed with the exceptions of peak level and downstream predictions in HN1 and HN2 flames but the downstream predictions are apparently better in the HN3 flame.

Scatter plots of mass fraction of H2O at axial locations x/D = 10 and x/D = 20 are shown in figure 5.47 for HN1, HN2 and HN3 flames. Once again, good predictions are observed with lower strain rate models. It should be noted that there are experimental uncertainties associated with the measurements of species concentrations but it is more significant in the minor species than for the major species.

The results presented in this chapter demonstrate that the flamelet models predictions based on the differential diffusion as well as with unity Lewis number are, in general, both qualitatively and quantitatively superior to those of other models. Overall predictions of major and minor species and flame temperatures, in all three flames are in reasonable agreement with experimental data. Results do, however, tend to unpredict mixture fractions, temperatures and species mass fractions in some axial and radial locations and in some cases. All these results point to the requirement for the further work to refine the models and to compare the results with other models and presumed form of probability density function used within the models and to investigate more accurately the averaged and fluctuating quantities for these flames.
Figure 5.43: Contours of mixture fractions; (a) HN1 flame, (b) HN2 flame, (c) HN3 flame. The flame front is at the location where \( Z = Z_{st} \).
Figure 5.44: Contours of temperatures; (a) HN1 flame, (b) HN2 flame, (c) temperature contours of HN3 flame with half side grid overlay.
Figure 5.45: Contours of mass fractions of H$_2$O; (a) HN1 flame, (b) HN2 flame, (c) HN3 flame.
Figure 5.46: Scatter plots of temperatures at (a) x/D = 10, (b) x/D = 20 for HN1 flame; (c) x/D = 10, (d) x/D = 20 for HN2 flame; (e) x/D = 10, (f) x/D = 20 for HN3 flame. Lines represent computed flamelet profiles with constant mass diffusivities and Le = 1.0. \( \alpha = 3.0/\text{s} \) (\( \chi_{\text{st}} = 0.64/\text{s} \)), \( \alpha = 100.0/\text{s} \) (\( \chi_{\text{st}} = 2.139/\text{s} \)), \( \alpha = 1800.0/\text{s} \) (\( \chi_{\text{st}} = 77.01/\text{s} \)).
Figure 5.47: Scatter plots of mass fraction of H$_2$O at (a) $x/D = 10$, (b) $x/D = 20$ for HN1 flame; (c) $x/D = 10$, (d) $x/D = 20$ for HN2 flame; (e) $x/D = 10$, (f) $x/D = 20$ for HN3 flame. Lines represent computed flamelet profiles with constant mass diffusivities and $Le = 1.0$. $\alpha = 3.0/\text{s} (\chi_{st} = 0.64 \text{/s})$, $\alpha = 100.0/\text{s} (\chi_{st} = 2.139 \text{/s})$, $\alpha = 1800.0/\text{s} (\chi_{st} = 77.01 \text{/s})$. 
5.9 Summary

1. The performance of the flame sheet, equilibrium and flamelet models was evaluated by comparing with the experimental data of H₂/N₂ fuels at different inlet velocities and compositions for jet diffusion flame configuration.

2. The laminar flamelet model provided better predictions than the other combustion models for predicting temperature, major and minor species.

3. The effect of the differential diffusion was studied and found to be less important for the prediction of H₂/N₂ flame. The unity Lewis number flamelet has provided a better representation of transport of the species.

4. The influence of different non-equilibrium parameters was evaluated and only the scalar dissipation rate was found to be suitable as a non-equilibrium parameter.

5. The modified k-ε model with $C_{ε1} = 1.60$ provides more accurate results and couplings of chemical models with turbulence were found well suited for the H₂/N₂ flames.

6. The increase of jet momentum results in longer flames observed from the stoichiometric mixture fraction contours.

7. The differences in the predicted results for different discretisation schemes were found to be very small.
Combustion/Radiation Modelling

The flame radiation plays an important role and being the dominant mode of heat transfer in fires and in many high temperature combustion systems, the classical studies concern mainly with the prediction of radiative heat transfer from a given flame to the surroundings. It is also important to study the influence of radiation heat transfer on the temperature and minor species but in most of the applications of flamelet models, however, radiation has not been accounted for as the assumption of adiabatic combustion is generally used (Marracino and Lentini (1997)). In most previous applications (Marracino and Lentini (1997), Bradley et al. (1998), Liew et al. (1984) and Rogg et al. (1986)) of flamelet models to turbulent flame predictions, neglecting the thermal radiation has lead to the overprediction of flame temperatures. Radiation heat transfer must therefore be accounted for when the laminar flamelet modelling approach is adopted to calculate turbulent flames. In some studies, however, radiative heat loss from the flame has been accounted by Jeng et al. (1982), Fairweather et al. (1992) and Young and Moss (1995) with the incorporation of a prescribed local heat loss fraction into the flamelet profiles without modeling the thermal radiation. A very few previous studies, such as Marracino and Lentini (1997) and Hossain et al. (2001) have included radiation using the diffusion-flamelet approach of methane-air flames.

The present chapter is concerned with the flamelet modelling of diffusion flames and coupling between the radiation heat transfer and flamelet model. Bray and Peters (1994) argued that when radiation heat loss is considered in a laminar flamelet, it implies that the radiation heat exchange occurs as thin gas radiative emissions to the surroundings within a thin region of high temperature. Moreover, radiations from actual flames are influenced by local properties as well as global effects arising from properties at distant locations. Bray and Peters (1994) suggested that to include radiation heat exchange into
the turbulent flame, emission and re-absorption over the whole range of length scales of the turbulent flame must be taken into account by solving a transport equation for enthalpy containing the radiation source term.

This study focuses, however, beyond the prediction of radiative flux from a flame; rather it aims to examine the active role of radiation in affecting combustion and turbulent processes of flames. To investigate the effect of radiation heat loss from the flames of consideration the flamelet model has been extended to introduce the effect of radiation heat transfer using the concept of enthalpy defect, as suggested by Bray and Peters (1994) and later used by Marracino and Lentini (1997) and by Hossain et al. (2001). Unfortunately, the detailed measured data of the radiative properties like radiative intensities and the corresponding wall heat fluxes for these flames are scant to compare with the predictions. However, the results presented here show that the predictive capability of the radiation adjusted model is better than that of adiabatic flamelet model.

6.1 Flamelet model with radiation

The flamelet equations constitute a parabolic set of coupled partial differential equations and as such the solution is governed by initial and boundary conditions and the time-dependent scalar dissipation rate. According to the laminar flamelet model, the thermochemical state of an adiabatic turbulent flame is completely determined by two parameters, the mixture fraction and the scalar dissipation rate at stoichiometric mixture fraction. Flamelet model is extended to introduce the effect of radiation heat transfer to predict the diffusion flames in consideration using the concept of enthalpy defect, as suggested by Bray and Peters (1994). The influence of radiation heat loss on the structure of laminar flamelet has been investigated by a number of researchers by including the radiation source term in the enthalpy equation of the counterflow diffusion flame or in the flamelet equations on the mixture fraction space (Chen et al. 1993; Chan et al. 1995; Balthasar et al. 1996). However, Bray and Peters (1994) argue that when the radiation heat loss term is retained in the flamelet equations, it implies that the radiation heat exchange occurs as thin gas radiative emissions to the surroundings within a thin region of high temperature. Radiation heat exchange is a global phenomenon and therefore radiation from practical flames does not correspond
to the radiation from laminar flamelets. To account for radiation heat loss, the flamelet library with the desired enthalpy defects corresponding to situations in turbulent flames can be generated by modifying the source term of the flamelet equation for enthalpy or simply by modifying the available enthalpy as suggested by Bray and Peters (1994). This approach has been successfully implemented by Marracino and Lentini (1997) and later by Hossain et al. (2001). A similar model for the inclusion of radiation heat transfer in the flamelet modelling has been reported by Young and Moss (1995) and Brookes and Moss (1999), which further accounts for soot radiation, however, this model cannot account for the finite rate chemistry. The method proposed by Bray and Peters (1994) which has been implemented by Marracino and Lentini (1997) and Hossain et al. (2001) accounts for the finite rate chemistry effects and hence, is implemented in this study. The present study gives a further insight into the flamelet radiation modelling by employing the ray tracing based discrete transfer method (DTM) of Lockwood and Shah (1981) to account for the radiation source term.

To include the nonadiabatic effect such as radiation heat loss, enthalpy defect of a flame, can be introduced as an additional flamelet parameter. Enthalpy defect, defined as the difference between the actual enthalpy and the adiabatic enthalpy of a flame and is given by:

$$\zeta(Z) = \Delta h = h - [h_0 + Z(h_f - h_0)]$$  \hspace{1cm} (6.1)

The scalar variable $\phi$ in a radiating flame now depends on the mixture fraction, the scalar dissipation rate and the enthalpy defect and takes the form:

$$\phi = \phi(Z, \chi_{st}, \Delta h)$$  \hspace{1cm} (6.2)

The average values of the scalar variables in the turbulent field can be recovered by introducing a joint probability density function (pdf) of mixture fraction and scalar dissipation rate as:

$$\bar{\phi} = \int_0^\infty \int_0^1 \phi(Z, \chi_{st}, \Delta h)P(Z, \chi_{st})dZd\chi_{st}$$  \hspace{1cm} (6.3)

where the effect of turbulent fluctuations of enthalpy are neglected according to Bray and Peters (1994) and Marracino and Lentini (1997). The assumption of statistical
independence between the mixture fraction and scalar dissipation rate leads to \( P(Z, \chi) = P(Z)P(\chi) \). The pdfs of the mixture fraction and the scalar dissipation rate are assumed to be a beta function and a log-normal function respectively (Peters, 1994). This model requires an input of a set of flamelet profiles with enthalpy defect as one of the parameters. According to the method described by Bray and Peters (1994), the flamelet profiles are organized as ‘shelves’. Each shelf represents a different value of the enthalpy defect, \( \Delta h \) and contains entries for the flamelet profiles. The flamelet libraries were generated for values of the scalar dissipation rate ranging from very low value close to chemical equilibrium to a large value close to extinction limit plus the inert state.

In the CFD code the mean enthalpy is calculated from its conservation equation, which contains the radiation source term \( S_h \):

\[
\frac{\partial}{\partial x_j} (\rho \hat{u}_j \hat{h}) = \frac{\partial}{\partial x_j} \left( \frac{\mu_{\text{eff}}}{\sigma_h} \frac{\partial \hat{h}}{\partial x_j} \right) + S_h
\]  

(6.4)

The radiation source term \( S_h \) is calculated from the discrete transfer model of radiation by Lockwood and Shah (1981). Once the value of mean enthalpy is obtained from the equation (6.4), the value of mean enthalpy defect is calculated at each grid node from Equation (6.1). The mean value of the scalar variables are then calculated from integration over the joint probability density function of mixture fraction and scalar dissipation rate and interpolation between the two consecutive enthalpy defect entries. Interpolation is required to calculate mean scalar variables, as only a limited number of shelves are available in the library. A linear interpolation is employed as given by Marracino and Lentini (1997):

\[
\bar{\phi} = \frac{\Delta \hat{h} - \Delta h_m}{\Delta h_{m-1} - \Delta h_m} \bar{\phi}_{m-1} + \frac{\Delta h_{m-1} - \Delta \hat{h}}{\Delta h_{m-1} - \Delta h_m} \bar{\phi}_m
\]  

(6.5)

where \( m \) denotes the library shelf index such that the enthalpy defect at a node lies between \( m-1 \) and \( m \), and where \( \bar{\phi}_m \) stands for:

\[
\bar{\phi}_m = \int_0^\infty \int_0^1 \phi(Z, \chi, \Delta h_m)P(Z, \chi, \Delta h) dZ d\chi
\]  

(6.6)
and $\phi_{m-1}$ may have a similar expression.

### 6.2 Flamelet library with enthalpy defect

To account for radiation heat loss, the flamelet library with enthalpy defect can be generated by modifying the source term in the flamelet equation for enthalpy or simply by modifying the available enthalpy according to Bray and Peters (1994). Marracino and Lentini (1997) described their method for the generation of flamelet library, where they have solved the flamelet equations in the mixture fraction space and reduced the temperature at the boundaries to achieve the required enthalpy defect. However to keep the boundary temperatures at a realistic level for the case of higher enthalpy defects, they have reduced the solution domain from the usual domain of $Z = 0.0$ to $Z = 1.0$ to a smaller range (for example, $Z = 0.01$ to $Z = 0.99$).

In the present study, the method proposed by Hossain et al. (2001) for generating flamelet library with enthalpy defect has been used to solve the problem of consideration. Instead of solving the flamelet equations in the mixture fraction space as Marracino and Lentini (1997), this method is based on the calculation of counterflow diffusion flame and is explained in details by Hossain et al. (2001). Instead of solving the flamelet equations in the mixture fraction space as Marracino and Lentini (1997), this method is based on the calculation of counterflow diffusion flame. In this method, an adiabatic flamelet library is calculated first for a given scalar dissipation rate. Figure 6.1 shows a schematic drawing of a flamelet temperature profile of this kind. To obtain a flamelet library with enthalpy defect, the temperature at the boundary is reduced from the adiabatic condition. This leads to unrealistically low boundary temperature for higher enthalpy defect which is avoided by feeding slightly rich air (corresponding to mixture fraction, $Z_1$) and slightly lean fuel (corresponding to mixture fraction, $Z_2$) to the counterflow diffusion flame boundaries. The flamelet profile for temperature then can be viewed as a downward shift from the adiabatic profile when the boundary temperatures are reduced by $\Delta T = \Delta h/C_p$ from the adiabatic condition. The flamelet profile with enthalpy defect then span between the mixture fraction $Z = Z_1$ and $Z = Z_2$ instead of $Z = 0$ and $Z = 1$. The $Z_1$ and $Z_2$ are the locations of mixture fraction close to 0 and 1 at which the new flamelet temperature profile intersects the $Z$-axis.
A small error is introduced in the integration of equation (6.3) for this reason but is expected to have minimal impact on the turbulent flame computation. The species concentrations corresponding to $Z_1$ and $Z_2$ of the adiabatic flamelet and temperatures $T(Z_1)-\Delta T_1$ and $T(Z_2)-\Delta T_2$ are then set as boundary conditions for the solution of counterflow diffusion flame in the new computation.

### 6.3 Discrete transfer radiation method

The discrete transfer (DT) method of Lockwood and Shah (1981) is a widely used calculation technique for radiation in combusting flows. The discrete transfer method based on the calculation of the radiative transfer along a number of predetermined ray paths using a recurrence relation obtained from solution of the differential equation of radiative transport. The radiative intensity entering and leaving each cell along this path is calculated for each ray, enabling the cell source term to be determined. The
calculation procedure takes into account all the local temperature and gas property values along a ray path, the method therefore, incorporates not only the local properties but also the global effects arising from properties at distant locations.

- The main assumption of the DT method is that the radiation leaving the surface element in a certain range of solid angles can be approximated by a single ray.

- The primary advantages of the DT method are threefold: it is a relatively simple model, the accuracy can be increased by increasing the number of rays, and it applies to a wide range of optical thicknesses.

The equation for the change of radiant intensity, $dI$, along a path, $ds$, can be written as:

$$\frac{dI}{ds} = k_g \left( \frac{E_g}{\pi} - I \right)$$

(6.7)

where $I$ is the intensity of radiation in the direction of $\Omega$, $k_g$ is the gas absorption coefficient, and $E_g$ is the black body emissive power, $\sigma T^4$ for $\sigma$ is the Stefan-Boltzmann constant. This equation can be integrated to yield the solution, $I(s)$ as:

$$I(s) = E_g (1.0 - e^{-k_g s}) + I_0 e^{-k_g s}$$

(6.8)

where $I_0$ is the radiant intensity at the start of the incremental path, which is determined by the appropriate boundary condition. The energy source in the gas due to radiation is then computed by summing the change in intensity along the path of each ray that is traced through the fluid control volume. The "ray tracing" technique used in the DT model can provide a prediction of radiation heat transfer between surfaces without explicit view-factor calculations. The corresponding recurrence relation of the solution (6.8) may be written as:

$$I_{n+1} = E_g (1.0 - e^{-k_g \delta s}) + I_n e^{-k_g \delta s}$$

(6.9)

where $n$ and $n+1$ denote successive locations separated by $\delta s$. The recurrence relation is applied along an arbitrarily chosen 'representative direction' $\Omega$'s, normally of equal angular spacing, emanating from arbitrarily chosen wall locations, usually the centre
points of grid cell surfaces coincident with a wall and terminating at the impingement locations on opposite walls. The heat balance at the wall is given by:

\[ q_{w,+} = (1.0 - \varepsilon_w)q_{w,-} + \varepsilon_w E_w \]  

(6.10)

The radiation intensity leaving the wall is:

\[ I_0 = \frac{q_{w,+}}{\pi} \]  

(6.11)

The incident flux is obtained from:

\[ q_{w,-} = \sum_{all} I_j (\Omega_j \cdot \vec{n}) \delta\Omega_j \]  

(6.12)

Where \( I_j \) is the calculated intensity in the discretised angle \( \delta\Omega_j \) and \( \vec{n} \) is the unit vector normal to the wall.

The radiation source term for the cell for each representative direction is given by:

\[ S_h = \sum_{all} (I_{n+1} - I_n) \delta A \delta\Omega \]  

(6.13)

At a wall location, usually the centre point of a grid cell surface coinciding with the wall, the representative direction is selected. Each representative direction is then traced through the cells until the far wall is reached. There, the intensity \( I_0 \) leaving the surface is taken to be the intensity of the adjacent centre point of the cell, where it is known. This process is repeated until the difference between the old negative flux and the new one is within some prescribed error. The weighted sum of the grey gases (WSGG) approach is used in the calculation of the emissivities and the absorption coefficients.

The global emissivity of the participating non grey gas mixture is represented by (Hottel and Sarofim 1967):

\[ \varepsilon_g = \sum_n a_{g,n}(T) \left[ 1.0 - e^{-k_{g,n}(p_w + p_c)L} \right] \]  

(6.14)

where the summation \( n \) is over the gases of the assumed mixture; \( p_w \) and \( p_c \) is the partial pressure of water vapour and carbon dioxide and \( L \) is the mean radiation path length.

The influence of temperature is introduced by the weighting co-efficient:
\[ a_{g,n} = b_{1,n} + b_{2,n}T \]  

(6.15)

These coefficients have been fitted to the gas mixture total emittance evaluated from the spectral data by Truelove (1976). The value of \( k_g \) required for the calculations is then obtained from the 'pseudo gray' approximation:

\[ E_g = 1 - e^{-k_g L} \]  

(6.16)

In this method the ray paths are calculated and stored prior to the fluid flow calculation. At each radiating face, rays are fired at discrete values of the zenith (polar) and azimuthal angles. To cover the radiating hemisphere, the zenith angle \( \theta \) is varied from 0 to \( \pi/2 \) and azimuthal angle, \( \phi \) from 0 to \( 2\pi \). Each ray is then traced to determine the control volumes it intercepts as well as its length within each control volume. The accuracy of the model depends mainly on the number of rays traced and the computational grid.

### 6.4 Radiation modelling of CO/H\(_2\)/N\(_2\) jet flame

The fuel considered here consists of 40% CO, 30% H\(_2\) and 30% N\(_2\) by volume and has a jet velocity of 76.0 m/s exits from 4.58-mm diameter nozzle with \( \text{Re}=16700 \). The jet nozzle is centred at the exit of a vertical wind tunnel; into a coflowing air stream of velocity 0.75m/s. Calculated temperatures and concentrations are given in Favre averaged values. The mean values of mixture fraction, mass fractions and temperature along the axis cross-section profiles at \((x/D=20), (x/D=30), (x/D=40)\) and \((x/D=60)\) are presented. Six shelves of flamelet profiles are generated corresponding to enthalpy defects, \( \zeta (=\Delta h) \) of 0.0, -15.0, -30.0, -40.0, -50.0 and - 60.0 kJ/kg and each shelf contains six flamelet profiles. The value of the extinction limit is different for different shelves and the extinction limit is reduced at higher enthalpy defects. This is quite obvious because the lesser amount of energy is available to sustain the flame at higher enthalpy defects and as a result the flamelet will be extinguished at a lower scalar dissipation rate. The flamelet library of temperature profiles for different enthalpy defects are shown in Fig.6.2. The flamelet library also includes similar profiles for density and mass fraction of species. As expected, the temperature is reduced by both
the scalar dissipation rate and the enthalpy defect. Another important feature is that the temperature profiles do not span the $Z=0$ to $Z=1.0$ range at higher enthalpy defects. This is because, slightly rich air and lean fuel are fed at the boundaries and the temperatures are reduced from the adiabatic condition to achieve the required enthalpy defects. The generation of flamelet library with enthalpy defects is explained in section 6.2.

The axial temperature profile is shown in Fig. 6.3. The effect of radiation is negligible until $x/D=25$. The effect of radiation becomes important in the high temperature zone of the flame and further downstream as well. The temperature with radiation model is about 50 K less than the temperature without the radiation effect.

Figure 6.4 shows contours of mean temperatures predicted using (a) adiabatic and (b) radiation adjusted flamelets. It can be observed that little difference between two approaches is evident in the vicinity of the near exit region of jet. However, further downstream, the inclusion of radiation heat loss is found to have significant effect on temperature patterns. The effect of radiation heat loss is found very small until $x/D=20$. The peak temperature is confined between $x/D=30$ and $x/D=54$. It can also be noted from the figures that the maximum radiation heat exchange occurring not at the peak temperature region, but further downstream.

Figure 6.5 represents the radial profiles of mean mixture fractions and its variances at four axial locations. Comparisons between the predicted radial distributions of mixture fractions and its variances for flamelet combustion model with unity Lewis number (solid lines) and flamelet model with differential diffusion (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 little differences between the differential diffusion and unity Lewis number flamelet models and all other computations considering enthalpy defect as a third parameter are performed with the unity Lewis number assumptions. The overall agreements between the mixture fraction predictions and the measurements are reasonably good by both the flamelet models with slight overpredictions for all the axial locations. Similar results have been observed in the predictions of mixture fraction variances.

Figure 6.6 shows the radial profiles of temperature and mass fraction of $H_2$ with and without radiation heat loss along with measurements at four axial locations. At the axial location $X/D=20$, the adiabatic flamelet model overpredicted the temperature levels
whereas the results obtained from the flamelet model with enthalpy defect are in good agreement with the experimental data. At X/D=30, both the models overpredicted the temperature profiles but the nonadiabatic model predicted better and results are closer to the measurements. Although both the model overpredicted the temperature in the decaying region, the predicted temperature profiles have shown rapid decay from the maximum position similar to the experimental data. At the axial location X/D=40 and further downstream station X/D=60, the measured temperature profiles tend to flatten and peak temperatures are located on the axis, which have been predicted well by radiation adjusted flamelet model but the adiabatic model significantly overestimated the levels of temperature. In general, it is observed that adiabatic model significantly overpredicts the measured levels of temperature at all of the four locations and the predictions of the radiation adjusted model are, in fairly good agreement with measurements over the whole length of the flame. As shown in Fig. 6.6 (b), the predicted mass fractions of H2 at X/D = 20 are in reasonably good agreement with the measurements with slight overpredictions at and around the jet axis. There are no significant differences found between the results obtained from adiabatic and radiation adjusted flamelet models at this axial location. At X/D = 30 and further downstream at X/D = 40 and 60, predictions of species mass fractions of H2 are well but very little amount of overpredictions found at and around the jet axis and in the decaying core zone for X/D = 30 and 40.

Figure 6.7 shows comparisons between the predicted radial distributions of the mass fractions of N2 and O2, along with the measurements at four axial stations. The first measurement station, at X/D = 20, the flamelet models with enthalpy defect predicted the N2 mass fraction near the jet axis with slight underprediction and shows overpredicted results for far axis region with gradual increase between these two region but the adiabatic model somehow performs better in this occasion. At X/D = 30 and 40 the nonadiabatic model predictions of N2 mass fractions are better in the near axis region and in the far axis region adiabatic model predicted better with slight overprediction. At X/D = 60 predictions by both the models are reasonably good with slight differences in results obtained from two models. In the predictions of mass fractions of O2, both the models predicted well at the first two measurement locations X/D = 20 and 30. For the last two axial measurement stations X/D = 40 and 60 models predictions of O2 mass fractions are fairly good with slight overprediction.
Comparisons between the predicted radial distributions of the mass fractions of CO$_2$ and CO, along with the measurements at four axial stations are shown in Fig. 6.8. The first measurement station, at X/D = 20, both the flamelet models with and without enthalpy defect underpredicted the peak value of CO$_2$ mass fraction and at the radial location which is little bit away from jet axis than the measured location. Predictions are observed better in the near axis and in the decaying regions but overpredicted results found for the far axis region provided by both the models. At X/D = 30 and 40 both the models predicted the CO$_2$ mass fractions with slight underpredictions at and about the jet axis and good predictions have been observed in the decaying regions. At X/D = 60, underpredicted results are found by both the models but adiabatic flamelet model provided comparatively better results of the CO$_2$ mass fractions. In the predictions of mass fractions of CO in Fig. 6.8 (b), both the models overpredicted the CO levels at and around the jet axis for the first three measurement locations X/D = 20, 30 and 40. The results are found more close to the measurements with slight overprediction in the decaying regions for these three locations. For the last axial measurement station X/D = 60 models predictions of CO mass fractions are reasonably good with very little differences in results obtained from two models. The overall performances of nonadiabatic flamelet model in the predictions of CO mass fraction are found better.

Figure 6.9 shows comparisons between the predicted radial distributions of the mass fractions of H$_2$O and OH, along with the measurements at four axial stations. The first measurement station, at X/D = 20, both the models underpredicted the mass fraction of H$_2$O near the jet axis towards the peak value and overpredicted in the decaying region. The peak value underpredicted by both the flamelet models at the radial location slight away from jet axis than the measured location. Similar results have been found for the measurement station X/D = 30. At X/D = 40, better predictions are provided by nonadiabatic flamelet model in the predictions of the species mass fractions of H$_2$O but slight underpredicted results provided by adiabatic flamelet model at and around the jet axis and in the decaying region. Both the flamelet models predicted the mass fractions of H$_2$O well at X/D = 60 but adiabatic model underpredicted slightly in the near axis region.
In the predictions of mass fractions of OH in Fig. 6.9 (b), Radical OH is formed through the rapid two-body reaction \( H + O_2 = OH + O \). The OH concentration decays towards equilibrium via the slower three-body recombination reaction \( H + OH + M = H_2O + M \) according to Barlow et al. (1990) and Drake and Blint (1988). In turbulent combusting flows, the rate of mixing is much slower than the chemical reaction rate of the two-body reaction, but much faster than that of the three-body reaction (Barlow et al., 1990). This has resulted in superequilibrium amount of OH in the flame as described by Dally et al. (1998a) in their study. The present computation shows that the flamelet model is able to predict the superequilibrium level of OH well. At \( X/D = 20 \) the nonadiabatic flamelet model predicted the OH level well at and around the jet axis towards the maximum value and overpredictions observed in the decaying region. There are significant level of overpredictions in the results obtained by adiabatic flamelet model for all the axial locations. At \( X/D = 30 \) the nonadiabatic flamelet model predicted the OH level with slight overpredictions at and about the jet axis and in the decaying region but closer to the measurements than those of \( X/D = 20 \). The nonadiabatic flamelet model predicted the OH level at \( X/D = 40 \) reasonably well with little bit overpredictions at the axis and in the decaying region around the jet axis.

For the last axial measurement station \( X/D = 60 \) models predictions of OH mass fractions are very good with little differences in the results obtained by the adiabatic and radiation adjusted flamelet models. The overall performances of nonadiabatic flamelet model in the predictions of OH mass fraction are found much better than the adiabatic flamelet model. The levels of OH are likely to be decreased with the increased enthalpy defect due to the reduction of reaction rate of \( H + O_2 = OH + O \) at lower temperature. Thus the results presented here show that the inclusion of radiation heat transfer remarkably improves the agreement between the prediction and the measurements.
Figure 6.2: Flamelet profiles of temperature with (a) ζ = 0.0 kJ/kg; (b) ζ = -15.0 kJ/kg; (c) ζ = -30.0 kJ/kg (d) ζ = -40.0 kJ/kg; (e) ζ = -50.0 kJ/kg; (f) ζ =
Figure 6.3: Axial temperature profiles along the centerline. —— without radiation; --- with radiation; ■ measurements

Figure 6.4: Predicted contours of temperatures, (a) without radiation heat transfer and (b) with radiation heat transfer.
Figure 6.5: Radial profiles of mean mixture fraction and mixture fraction variance in the CO/H2/N2 flame experimental (EXP), flamelet model with unity Lewis number (FLU), flamelet model with differential diffusion (FLD)
Figure 6.6: Radial profiles of (a) temperature and (b) mass fraction of H₂ in the CO/H₂/N₂ flame. Experimental (EXP), adiabatic flamelet model (ADB), flamelet model with radiation (RAD)
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Figure 6.7: Radial profiles of mass fractions of (a) N₂ and (b) O₂ in the CO/H₂/N₂ flame. Experimental (EXP); adiabatic flamelet model (ADB); flamelet model with radiation (RAD)
Figure 6.8: Radial profiles of mass fractions of (a) CO\textsubscript{2} and (b) CO in the CO/H\textsubscript{2}/N\textsubscript{2} flame. Experimental (EXP); adiabatic flamelet model (ADB); flamelet model with radiation (RAD).
Figure 6.9: Radial profiles of mass fraction of (a) H$_2$O and (b) OH in the CO/H$_2$/N$_2$ flame. Experimental (EXP), adiabatic flamelet model (ADB), flamelet model with radiation (RAD).
6.5 Radiation modelling of H$_2$/N$_2$ jet flame

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 which is HN2 flame as described in table 5.1. The simulated results have been compared in figures 6.10-6.13 along with the experimental data. The computed and measured mixture fraction and its variance, temperature and species mass fractions profiles have been presented at six axial cross-sections x/D=5, x/D=10, x/D=20, x/D=40, x/D=60 and x/D=80.

The control parameters of the flamelet are the mixture fraction, Z, the strain rate, $\alpha$ and the enthalpy defect $\Delta h$. 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. Figure 6.10 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 all other computations considering enthalpy defect as a third parameter are performed 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 of 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 6.11 shows the computed radial profiles of temperature with and without radiation heat loss along with measurements at six near and far-field axial locations. At the axial location X/D=0.5, the computed mean temperature distribution obtained from both the models are in good agreement with the experimental data, and are qualitatively correct in predicting the influence of the onset of combustion. At X/D=10 and 20, combustion models predicted the temperature profiles well from the centreline towards the peak value. The adiabatic model overestimated the maximum temperature and both the model predicted the peak value little bit away from the centreline than that of measurements. Although both the model overpredicted the temperature in the decaying region, the predicted temperature profiles have shown rapid decay from the maximum position similar to the experimental data. At the axial location X/D=40 and far downstream station, X/D=60 and 80, the measured temperature profiles gradually flattening and peak temperatures are located on the axis, indicating a spreading of the reaction zone and that the flame front has moved towards the axis. This trend is captured in the predictions obtained using the flamelet model with radiation, but the adiabatic model significantly overpredicted the levels of temperature. In general, it is observed that the adiabatic model generates good quality predictions in the near axis and near exit plane region but in the downstream region, this model significantly overpredicts the measured levels of temperature at and around the flame axis. In contrast, the predictions of the radiation adjusted model are, in good agreement with measurements over the whole length of the flame.

Figure 6.12 shows comparisons between the predicted radial distribution of the mass fraction of $\text{H}_2$ and $\text{H}_2\text{O}$, along with the measurements at six axial locations. The first measurement station, at X/D = 0.5, which is near exit region of the jet, where the predicted species mass fractions are in good agreement with the experimental data, indicating the onset of combustion is correctly predicted. At X/D = 10 and 20, which are located within the core of combusting zone, the predicted $\text{H}_2$ mass fractions are in reasonably good agreement with the data. Computations of $\text{H}_2\text{O}$ mass fractions are fairly good with slight overpredictions in near axis values and the peak value for X/D = 10. Some discrepancies exist in the prediction of $\text{H}_2\text{O}$ mass fractions at X/D = 40, in this case overpredicted results found for all over the reacting zone. $\text{H}_2$ predictions are
reasonably good at this location. Further downstream at X/D = 60 and 80, predictions of species mass fractions of H₂ and H₂O are well but slight overpredictions in H₂O found in the decaying core for X/D = 60 and around the jet axis for X/D = 80. All the predicted results for the mass fractions of H₂ and H₂O show little difference between the adiabatic and radiation adjusted flamelet models. Finally, Fig. 6.13 shows contours of mean temperatures predicted using adiabatic and radiation adjusted flamelets. As noted earlier, little difference between two approaches is evident in the vicinity of the near exit region of jet. However, the effects of radiation heat transfer are found to be more significant in the downstream levels. In particular, consideration of radiation heat transfer into the combustion model remarkably improves the agreement between predictions and the measurements specially, in the temperature profiles.
Figure 6.10: Radial profiles of mean mixture fractions (mf) and its variances (mfv) at six axial locations: ○ measurements; — flamelet model with differential diffusion; —— unity Lewis number flamelet model.
Figure 6.11: Radial profiles of temperatures at six axial locations: o measurements; —— adiabatic flamelet model; —— flamelet model with radiation.
Figure 6.12: Predicted and measured mass fractions of H₂ and H₂O: —— adiabatic flamelet model; – – – flamelet model with radiation; measurements (symbols).
6.6 Radiation modelling of Sydney HM2 flame

The Sydney HM2 flame simulated here obtained from the mixture of Hydrogen and Methane. The fuel consists of 50% CH₄ + 50% H₂ (by volume) and has a fuel jet velocity of $U_j = 178$ m/s with coflowing air velocity $U_e = 40$ m/s. The simulated results for different profiles have been compared in figures 6.14-6.21 along with the experimental data. The computed and measured mixture fraction and its rms values, temperature and species mass fractions profiles have been presented at six axial cross-sections $X=13, 30, 45, 65, 90$ and $120$ mm.

Figures 6.14 and 6.15 represent the radial profiles of mean mixture fractions and its rms values respectively at six axial locations. Comparisons between the predicted radial distributions of mixture fractions and its rms for flamelet model with differential diffusion (dashed lines) and unity Lewis number flamelet model (solid lines), along with the measurements at various near- and far-field axial stations. As can be observed from the figures, the predicted results show little differences between the differential diffusion and unity Lewis number flamelet models and as have done in the previous two cases all other computations considering enthalpy defect as a third parameter are performed 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 decaying regions for the first three measurement stations, $X=13, 30$ and $45$ mm. At $X = 65$ and $90$
mm, slight underpredicted results found at the jet axis and in the decaying region. At X=120 mm, although there are no adequate measured data to compare but the predicted mixture fraction results have shown good agreement with the available data. In the predictions of RMS of mixture fractions in Fig. 6.15, the predicted maximum variations have been found higher than that of measurements at the axial locations X = 13 and 30 mm. Also the peak values observed slightly away from the axis than that of the measured locations. Overpredictions found in the decaying region but in the far axis region the predicted results agreed well with the measurements with slight overprediction. At X = 45, 65 and 90 mm, the flamelet model with differential diffusion predicted the variations level well in the near axis region up to the maximum level, both the models have shown overpredictions in the decaying region and predicted well in the far axis region. At X=120, the flamelet model with differential diffusion predicted have shown better agreement with the available measurements. Six shelves of flamelet profiles are generated corresponding to enthalpy defects of 0.0, -25.0, -50.0, -75.0, -100.0 and -125.0 kJ/kg. Each shelf contains a single flamelet profile corresponding to the strain rate of a=100.0 /s. The predicted temperature with and without radiation is shown in Fig. 6.16. The effects of radiation heat transfer appear significant inside the recirculation zone of this flame as well as in the other locations. The reduction of temperatures are prominent at axial locations X=13 and 30 mm which are within the recirculation zone. At X=13 mm, the peak temperature is reduced by about 230 K due to radiation. The difference in temperature with and without radiation is reduced to about 190 K at X=30 mm. Adiabatic model overestimated the temperature level significantly at these axial locations whereas the agreement between the predictions of radiation adjusted flamelet model and experiments are much better at these locations. Further downstream at X= 45 and 65 mm, flamelet model with radiation predicted the temperature profiles well from the centreline towards the peak value and then both the model overpredicted the temperature in the decaying region but the predicted temperature profiles have shown rapid decay from the maximum position similar to the experimental data. At the axial location X=90mm and far downstream station, X=120mm, the temperature profiles gradually flattening and peak values of temperatures are closer to the axial values. This trend is captured in the predictions obtained from both of the flamelet models but the results obtained from radiation adjusted flamelet model are closer to the available experimental data and the adiabatic
model significantly overpredicted the levels of temperature. In general, it is observed that adiabatic model generates good quality predictions in capturing the trend and shape of the temperature profiles but this model significantly overpredicts the measured levels of temperature at all the locations whereas, the predictions of the radiation adjusted model are, in good agreement with measurements with some exceptions for the decaying region.

Comparisons between the predicted radial distribution of the mass fraction of H₂O and CO₂, along with the measurements at six axial locations have been shown in Figs. 6.17 and 6.18. The effects of radiation heat transfer on H₂O mass fractions are found very small as shown in the figures. The first two measurement stations, X = 13 and 30mm, which are within the recirculation zone, where both the models predicted the species mass fractions of H₂O well in the near axis region up to the maximum level. Then both the models overpredicted the H₂O mass fractions in the whole of the maximum valued flattened region and in the far axis region with slight better results from radiation adjusted flamelet model. At X = 45 mm, predicted results provided slightly higher values at jet axis and in the peak region of H₂O mass fractions. Good predictions have been found in all other near and far axis regions. At X = 65 and 90 mm, near and far axis predictions are well but overpredictions have been found in the decaying regions. At X=120mm, both the flamelet models have provided reasonable results and have shown better agreement with the available data.

In the prediction of mass fractions of CO₂ in Fig. 6.18, at the first measurement location X = 13 mm both the models predicted the mass fractions of CO₂ well in the near axis and decaying region but have provided overpredicted results in the maximum valued flattened region. At X= 30mm, predictions are well at the jet axis and very close to it but underprediction found in the increasing zone and overpredictions for the rest of the regions. At X = 45 and 65 mm, models predicted the values of decaying region well but overpredictions observed in the all other regions with comparatively better results provided by the flamelet model with radiation. At X = 90 mm, significant overpredictions found all over the flame zone but better agreement observed in the far axis region. Similar results observed at X=120mm in comparison with the available data.

Comparisons between the predicted radial distributions of the mass fractions of CO along with the measurements at six axial locations have been shown in Figs. 6.19. Even
if the effects of radiation heat transfer on mass fractions of CO are found very small, difference in results obtained from adiabatic and radiation adjusted flamelet models are significant at the axial location \( X = 13 \text{mm} \). At this location adiabatic model predicted better up to the maximum value and nonadiabatic model predicted better in the maximum valued flattened region, in the decaying and far axis region both the model predicted well with slight over prediction. The second measurement station, \( X = 30 \text{mm} \), both the models predicted the species mass fractions of CO well in the near axis region up to the maximum level then both the models provided slightly over predicted results in the decaying and far axis region. At \( X = 45, 65 \) and 90mm, overpredictions found in the decaying region and both the models provided good results in the far axis regions and slight higher values near the jet axis. At \( X=120 \text{mm} \), both the flamelet models have provided reasonable results and have shown overpredictions in comparison to the available data.

Radial distributions of the mass fractions of OH along with the measurements at six axial locations have been shown in Fig. 6.20. The incorporation of radiation heat transfer in the flamelet modelling is shown to improve the prediction of the radical species OH and at all the axial locations radiation adjusted flamelet model found to predict better than the adiabatic model. Since experimental measurements of O are not available, the prediction of OH radical can be treated as an indication of O prediction. At the axial location \( X = 13 \text{mm} \), both the adiabatic and radiation adjusted flamelet models predicted the mass fractions of OH well up to the maximum level starting from the jet axis but the peak value over predicted and observed little bit away from the centreline than that of the measurement. Further away, both the models provided almost same results with over prediction. The second measurement station, \( X = 30 \text{mm} \), both the models predicted the species mass fractions of OH well at the jet axis and in the near axis region. The maximum value predicted slightly closer to the jet axis than that of the measured location and then better predictions found in the decaying and far axis region. At \( X = 45, 65 \) and 90mm, predicted results are well enough at the jet axis and in the near and far axis region and overpredictions found in peak value and in the decaying region. At \( X=120 \text{mm} \), the flamelet models have provided reasonable results and have shown good agreement with the available measured data. Figure 6.21 represents the radial distributions of the mass fractions of nitric oxide (NO) along with the measurements at six axial stations. Thermal radiation from flames reduces the local
temperatures sufficiently to affect the production rate of NO and the incorporation of radiation heat transfer in combustion model is found to improve the prediction of the species NO significantly. At the axial location $X = 13\text{mm}$, the nonadiabatic flamelet model predicted the mass fractions of NO well with slight overprediction in the maximum valued flattened region whereas the adiabatic model overpredicted the NO level significantly at this location. The second measurement station, $X = 30\text{mm}$, both the models predicted the species mass fractions of NO well at the jet axis and in the near and far axis and in the decaying region. The maximum value overpredicted significantly by adiabatic model and radiation adjusted model provided good results closer to the measurements. At $X = 45\text{mm}$, predicted results are well near the jet axis and in the decaying and far axis region and overpredictions found in the region of positive gradient and in the peak value with better results provided by the nonadiabatic flamelet model. At $X = 65\text{mm}$, results obtained from radiation adjusted flamelet model have shown good agreement with the measured data with very little overprediction at the peak value whereas the adiabatic model have shown highly overpredicted results at and around the peak zone. At $X = 90\text{mm}$, nonadiabatic model predicted the mass fractions of NO well in the decaying and far axis region and slight overpredicted results provided in the near axis and peak values. The performances of adiabatic model in the far axis and decaying region are reasonable but higher rate of overpredictions observed in the near jet axis and peak valued zone. At $X=120\text{mm}$, the reasonable results obtained from the models predictions. At this axial location the radiation adjusted flamelet model provided slight overpredictions according to the available measured data near the axis and overall higher valued results provided by the adiabatic flamelet model. The effect of flame radiation on the mass fractions of NO in calculations of the jet flames is to reduce the predicted level of NO because of the sufficient reduction of local temperature due to radiation heat loss; the influence of radiation on major species mass fractions is not so significant as on the minor species. In particular, consideration of radiation heat transfer into the combustion model remarkably improves the agreement between predictions and the measurements specially, in the temperature profiles.
Figure 6.14: Radial profiles of mean mixture fraction in the HM2 flame. Experimental (EXP), flamelet model with unity Lewis number (FLU), flamelet model with differential diffusion (FLD)
Figure 6.15: RMS of mixture fraction the in the HM2 flame Experimental (EXP), flamelet model with unity Lewis number (FLU), flamelet model with differential diffusion (FLD)
Figure 6.16: Radial profiles of temperatures in the HM2 flame. Experimental (EXP), adiabatic flamelet model (ADB), flamelet model with radiation (RAD)
Figure 6.17: Radial profiles of mass fraction of H₂O in the HM2 flame. Experimental (EXP), adiabatic flamelet model (ADB), flamelet model with radiation (RAD)
Figure 6.18: Radial profiles of mass fraction of CO$_2$ in the HM2 flame. Experimental (EXP), adiabatic flamelet model (ADB), flamelet model with radiation (RAD).
Figure 6.19: Radial profiles of mass fraction of CO in the HM2 flame. Experimental (EXP), adiabatic flamelet model (ADB), flamelet model with radiation (RAD)
Figure 6.20: Radial profiles of mass fraction of OH in the HM2 flame. Experimental (EXP), adiabatic flamelet model (ADB), flamelet model with radiation (RAD).
Figure 6.21: Radial profiles of mass fraction of NO in the HM2 flame. Experimental (EXP), adiabatic flamelet model (ADB), flamelet model with radiation (RAD).


### 6.7 Discussion

The incorporation of radiation heat transfer in the laminar flamelet modelling is shown to improve the predictive capabilities of the model and remarkably improves the agreement between the prediction and the measurements specially in the temperature profiles. In general, it is observed that adiabatic model significantly overpredicts the measured levels of temperature in most of the cases and the predictions of the radiation adjusted model are, in fairly good agreement with measurements over the whole length of the flame. Since experimental measurements of O are not available, the prediction of OH radical can be treated as an indication of O prediction.

As the radiation heat transfer influences the temperature field, thermal radiation from flames reduces the local temperatures sufficiently to affect the production rate of NO and the incorporation of radiation heat transfer in combustion model is found to improve the prediction of the species NO significantly. Thus the present calculation method will be very helpful for improving the prediction of NO emission. However, the method for incorporating the radiation heat transfer implemented here is valid for the assumption of unity Lewis number of the flamelet. The flamelet with unity Lewis number on the other hand has yielded severe underprediction of NO. The flamelet with the differential diffusion effect results in a better prediction of NO, which will be discussed further in the next chapter. The current calculation method, therefore, has to be modified before the influence of radiation heat transfer on the emission of NO can be studied. The incorporation of radiation heat transfer with the differential diffusion flamelet is however very complex. The main problem arises in the generation of flamelet libraries with enthalpy defects. With the differential diffusion effect of the species, the enthalpy defect across the flamelet will not remain constant at those values imposed on the boundaries. Therefore, a single shelf can not be specified by a fixed enthalpy defect and the Eq. 6.1 will no longer be valid. If each shelf \( m \) is computed by enforcing the enthalpy defect \( \Delta h \) at the boundaries, the enthalpy defect across the flamelet will be a function of both \( Z \) and \( \chi_{st} \) (Marracino and Lentini 1997):
\[ \zeta(Z, \chi_{st})_m = [h(Z, \chi_{st})]_m - [h(Z, \chi_{st})]_a \]  
(6.17)

where index \( a \) refers to the shelf in adiabatic condition. This equation replaces the definition in Eq. 6.1. The incorporation then requires a very complex integration and interpolation than those given by Eqs. (6.5) and (6.6).

Some general features of the radiation heat exchange in the HM2 flame can be revealed from the contour plots of temperatures (a) without radiation and (b) with radiation heat loss in Fig. 6.22. It can be observed that little difference between two approaches is evident in the vicinity of the near exit region of jet and in the recirculation zone. However, further downstream, the inclusion of radiation heat loss is found to have significant effect on temperature patterns.

![Figure 6.22: Predicted contours of temperatures, (a) without radiation and (b) with radiation heat loss for HM2 flame](image)
The maximum radiation is confined at the high temperature zone inside the recirculation region. The effect of the radiation heat transfer on the chemical reaction is included through the enthalpy defect and the enthalpy in the flow field is influenced by the convection and diffusion processes as well. The combustion inside the recirculation zone are more influenced by radiation heat transfer which are significant in the downstream level as well.

The calculations reported here was carried out using 18x18 rays per cell. Further increase of the rays couldn’t improve the mean values significantly. Therefore it is concluded that in the present prediction process the radiation calculation using 18x18 rays is accurate enough to present predictions without imposing severe overheads on the overall calculation procedure.

Finally, the convergence speed of the coupled radiation-combustion calculation algorithm somehow depends on the frequency at which the radiation algorithm is called relative to the main CFD routine. Earlier in the calculation, a low frequency can be detrimental to the convergence as large perturbations are inflicted on the enthalpy equation (Bresslof et al. 1996). It is not essential to update the radiation field with every CFD iteration; rather such an approach is very demanding. As a compromise, the radiation routine is called after every 5(or 6) CFD iterations for the present analysis. However, it should be noted that no sensitivity analysis was carried out to determine the optimum frequency of iterations.

6.8 Summary

1. The importance of radiation heat loss in flamelet modelling of turbulent combustion has been demonstrated and a coupled flamelet/radiation modelling strategy is presented that combines the prediction of finite rate chemistry effects with a detailed solution of the radiative heat exchange.

2. The effect of the radiation heat transfer on temperature and mass fractions of major species and minor species like OH and NO are studied and comprehensive validation of CFD based flamelet/radiation results for reacting flows It is shown
that for the geometries presented here, the effect of radiation is important at regions where the temperatures are more than 2000 K.

3. The effects of the radiation are found very important for OH and NO predictions. The inclusion of radiation heat transfer into the simulation remarkably improves the prediction of OH mass fraction.

4. Thermal radiation from flames reduces the local temperatures sufficiently to affect the production rate of NO and the incorporation of radiation heat transfer in combustion model is found to improve the prediction of the species NO significantly.

5. The present study demonstrates that the radiation adjusted flamelet model for combustion leads to encouraging results that reproduce the overall characteristics of radiating turbulent diffusion flames and the flamelet/radiation modelling strategy presented here has potential for further improvement.
Modelling of NOx

Nitrogen present in the combustion system, provided either in the combustion air or the fuel, reacts during combustion to form NO and NOX, and also N2O in fairly minor amounts. In the last half of the twentieth century, it became apparent that nitric oxide (NO) and nitrogen dioxide (NO2), collectively called NOX, is a major contributor of photochemical smog and ozone in the urban air, more general the atmosphere (Seinfeld 1986). 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 NOX emitted. With the steady increase in combustion of hydrocarbon fuels, the products of combustion are distinctly identified as a severe source of environmental damage. The major combustion products are carbon dioxide and water. These products were, until recently, considered harmless. Now, even the carbon dioxide is becoming a significant source in the atmospheric balance, and concerns of a global greenhouse effect are being raised.

Furthermore, NOX participates in the chain reaction removing ozone from stratosphere with the consequence of increased ultraviolet radiation reaching the earth’s surface (Johnston 1992). Consequently, minimization of NOX production has become a most important topic in combustion. This minimization has been and continues to be achieved through increased comprehension of the interaction of the chemical kinetic mechanisms that generate NOX and understanding of the interaction of chemical kinetics and fluid dynamics. These models guide investigations toward new ways for the minimization of these pollutants.
7.1 Formation and emission of NOx

Strong dependence of the NOx 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 NOx. The amounts of NOx emitted from larger systems depend on the combustion hardware used (Sawyer and Starkman (1968)).

Considering NOx emissions by Emission Index (EI, milligrams of NOx per gram of fuel), spark-ignition engines yield EI = 16.3 whereas regenerative gas turbines reach EI = 13.5 and aircraft turbojets have EI = 5.5. Other practical factors have also been studied (Cunningham 1978): When burning residual fuel oils in boilers air preheat has considerable influence on NO formation, whereas oil preheat showed little influence on NO. Conditions that favour high combustion intensities yield relatively higher NO emissions (Nimmo et al. 1991), such as small droplet size (Sarv et al. 1983), narrow spray angle, etc.

The NOx concentration in the exhaust of an oil-fired boiler (Gills 1973) indicates that the NOx concentration decreases with excess air. Also the boiler size plays an important role on the concentration of NOx in the flue gases. Factors like the method of firing have little influence.

Maximum formation of NO occurs in gas turbines when the temperature is at its peak and at an equivalence ratio between 0.8 and 1.0 (Sawyer and Starkman 1968). The most important factor affecting NO formation in gas turbines is flame temperature (NOx \propto \exp(0.009 T)) (Gupta 1992); other important factors are residence time and oxygen concentration, and they are significant insofar as they affect flame temperature.

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

- 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 routes are now identified in the formation of NOx (Bowman 1993). These are the thermal route, the prompt route, the N₂O (nitrous oxide) route, and the fuel-bound nitrogen route.

7.1.1 Thermal NO (Zeldovich-NO)

Thermal NO formed by high-temperature oxidation of atmospheric nitrogen. The role of nitrogen contained in the air in NO formation was initially postulated by Zeldovich (Zeldovich, 1946). Thermal NO or Zeldovich-NO (after Zeldovich, 1947) is formed by the elementary reactions (Baulch et al. 1994)

<table>
<thead>
<tr>
<th>No.</th>
<th>Reaction</th>
<th>( B_\theta )</th>
<th>( a_\theta )</th>
<th>( E_\theta )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>( \text{N}_2 + \text{O} = \text{NO} + \text{N} )</td>
<td>1.900E+14</td>
<td>0.0</td>
<td>318.7</td>
</tr>
<tr>
<td>2</td>
<td>( \text{O} + \text{NO} = \text{N} + \text{O}_2 )</td>
<td>2.400E+09</td>
<td>1.0</td>
<td>161.6</td>
</tr>
<tr>
<td>3</td>
<td>( \text{NO} + \text{H} = \text{N} + \text{OH} )</td>
<td>1.300E+14</td>
<td>0.0</td>
<td>205.7</td>
</tr>
</tbody>
</table>

The name "thermal" is used, because the first reaction has a very high activation energy due to the strong triple bond in the \( \text{N}_2 \)-molecule, and is thus sufficiently fast only at high temperatures. This route is enhanced by the presence of super-equilibrium O and OH, and is very sensitive to temperature. Because of its small rate, reaction 1 is the rate limiting step of the thermal NO-formation. For this reason thermal-NO\(_x\) shows a strong exponential dependence on temperature. The contribution of thermal-NO to the total NO formation is small below 1,370 °C (Gupta 1992), but becomes very important above 1,400 °C. Thus the peak flame or combustion temperatures are used as an indication of the importance of thermal-NO.

Other factors which also affect NO\(_x\) formation are fuel/air mixing processes (related to local levels of excess air), combustion intensity and pre-heating of the combustion air. Thermal-NO has also been shown to increase linearly with residence time. Several mathematical expressions have been derived to estimate the rate of thermal-NO formation in combustion systems. If the processes leading to the formation of thermal-NO\(_x\) occurred long after those of combustion, the amounts of NO could be calculated from the equilibrium conditions (Bowman 1979). Thus assuming a steady-state
approximation for the N atom concentration, the formation rate of NO would involve the knowledge of the local temperature and the concentrations of O₂, N₂ and OH only, these being obtained from the post-combustion conditions. The following expression shows the large dependence of NO formation on temperature and oxygen concentration:

\[
\frac{d}{dt}[\text{NO}] = 6 \times 10^{16} \exp \left( -\frac{69,090}{T_{\text{eq}}} \right) \sqrt{\frac{[\text{O}_2]_{\text{eq}}}{T_{\text{eq}}}} [\text{N}_2]_{\text{eq}}
\] (7.1)

Toof (1986) found the residence time for NO formation in gas turbines to be proportional to the velocity of the air jet entering the combustor (V), the diameter of the combustor (D), and also to a function of the amount of excess air (λ):

\[
t = \frac{0.161D}{V(0.5+0.22[\lambda-1])}
\] (7.2)

Another attempt to estimate the formation of NO in gas turbines was given by Sawyer and Starkman (1968). Based only on the reaction 1 in table 7.1, where the O atom concentration is fixed by the equilibrium:

\[ \text{O}_2 \rightleftharpoons \text{O} + \text{O} \] (7.3)

and assuming that the level of NO is fixed by the kinetics of formation rather than from equilibrium formation in the primary zone, they proposed the following expression:

\[
\frac{[dX_{\text{NO}}]}{dt} \propto \sqrt{P} \cdot \exp \left( \frac{-133800}{RT} \right)
\] (7.4)

It shows a very strong dependence on temperature and inverse dependence on pressure, which are consistent with experimental results. The reverse reactions which would result in conversion of NO back into N₂ and O₂ are relatively slow in gas turbines (Starkman et al. (1971)). Thus, NO, once formed, does not decrease in concentration at an appreciable rate by cooling.
7.1.2 Prompt NO (Fenimore-NO)

The prompt NO formed by reaction of \( \text{N}_2 \) with hydrocarbon fractions. The mechanism of prompt or Fenimore NO was postulated by Fenimore (1979), who measured NO above a hydrocarbon flat flame and noted that the NO did not approach zero as the probe approached the flame from the downstream side, as the Zeldovich mechanism predicts. The additional mechanism that is promptly producing NO at the flame front is more complicated than thermal NO, because the prompt NO results from the radical CH, which was previously considered to be an unimportant transient species that is generated through a complex reaction scheme. The CH, which is formed as an intermediate at the flame front only reacts with the nitrogen of air, forming hydrocyanic acid (HCN), which reacts further to NO.

\[
\text{CH} + \text{N}_2 \rightarrow \text{HCN} + \text{N} \rightarrow \cdots \rightarrow \text{NO} \\
\text{CH} + \text{N}_2 \rightarrow \text{HCN} + \text{N} \rightarrow \cdots \rightarrow \text{N}_2
\]

Precise information about the rate-limiting step CH\( + \text{N}_2 \rightarrow \text{HCN} + \text{N} \) is rather rare in the literature, as can be seen from the Arrhenius plot of the rate coefficient.

7.1.3 NO Generation via Nitrous Oxide (N\(_2\)O)

The nitrous oxide (N\(_2\)O) mechanism is analogous to the thermal mechanism in that O-atom attacks molecular nitrogen. However with the presence of a third molecule \( M \), the outcome of this reaction is N\(_2\)O (postulates first by Wolfrum (1972)),

\[
\text{N}_2 + \text{O} + \text{M} \rightarrow \text{N}_2\text{O} + \text{M}.
\]

The N\(_2\)O may subsequently react with O-atoms to form NO (Malte and Pratt 1974),

\[
\text{N}_2\text{O} + \text{O} \rightarrow \text{NO} + \text{NO} \quad E_a = 97 \text{ kJ/mol}.
\]

This reaction has been often overlooked since it usually is an insignificant contributor to the total NO. However, lean conditions can suppress the formation of CH and, hence, lead to less Fenimore NO, and low temperatures can suppress the Zeldovich NO. What remains is NO generated via N\(_2\)O, which is promptly at high pressures because of the
three-body reaction and, typical for three-body reaction, has allow activation energy so that low temperatures do not penalize this reaction as much as they do the Zeldovich-NO reaction. All these circumstances lead to the N₂O route being the major source of NO in lean premixed combustion in gas turbine engines (Correa 1992).

7.1.4 Fuel NO (Conversion of Fuel Nitrogen into NO)

It forms due to oxidation of fuel-bound nitrogen compounds. The conversion of fuel-nitrogen, sometimes called fuel-bound nitrogen (FBN), into NO is mainly observed in coal combustion, because even "clean" coal contains about 1% chemically bound nitrogen by mass. The nitrogen-containing compounds evaporate during the gasification process and lead to NO formation in the gas phase.

The conversion of the nitrogen-containing compounds into NH₃ (amonia) and HCN (hydrocyanic acid) is usually quite fast and, thus not rate-limiting. The rate-limiting steps are the reactions of the N-atoms. In fuel-lean atmospheres HCN reacts to form NH:

\[
\text{HCN} + \text{O} \rightleftharpoons \text{NCO} + \text{H}
\]

\[
\text{NCO} + \text{HO} \rightleftharpoons \text{NH} + \text{CO}
\]

whereas in fuel-rich systems the mechanism is through NH₂:

\[
\text{HCN} + \text{OH} \rightleftharpoons \text{HNCO} + \text{H}
\]

\[
\text{HNCO} + \text{H} \rightleftharpoons \text{NH}_2 + \text{CO}
\]

\[
\text{NH}_2 + \text{H} \rightleftharpoons \text{NH} + \text{H}_2
\]

The subsequent oxidation of the amine species is a rapid process which occurs via two routes, depending on the availability of oxidant in the combustion environment (Toof 1986). The relative contribution from different pathways depends on fuel type, temperature, pressure and residence time (Dally et al. (1996)). 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).

7.2 Flamelet modelling of NOx

The prediction of thermal NO by the laminar flamelet model for the H₂/N₂ flame and Sydney HM1 and HM3 flames are presented here. The flamelet model describes the non-equilibrium chemistry by introducing the scalar dissipation rate as a parameter to account for non-equilibrium effects which has been described in the previous chapters. However, the flamelet approach still relies on the assumption that the time scales for chemical kinetics are much shorter than the time scales for convection and diffusion. With this concept, the mean temperature, density and composition in the turbulent field is obtained by appropriately averaging scalar variable versus mixture fraction profiles of the flamelets, each of the flamelets is subjected to different level of fluid dynamic stretching characterised by the scalar dissipation rate. Combustion generated pollutants such as NO on the other hand can not be obtained from the flamelet library as the reaction rate of NO formation is slow; it is kinetically limited instead of diffusion limited. Therefore, an averaged transport equation for the mean NO-mass fraction, $\bar{Y}_{NO}$, has to be solved. For a given $(Z, \chi)$ state, the source term for NO is evaluated from the corresponding chemistry states of major and minor species from the flamelet library. It is also essential that the existing laminar flamelet model is assessed against a large number of nonintrusive, spontaneous measurements to understand different factors influencing the NO formation before the predictive capability can be confidently implemented for design purposes.

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.
where $\sigma_{\text{NO}}$ is the turbulent Schmidt number, which is taken as 0.7. For a given $(f, \chi)$ 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

$$\dot{\omega}_{\text{NO}} = \dot{S}_{\text{NO}} - \frac{[\text{NO}]}{\tau_{d\text{-NO}}}$$

(7.6)

Where the first term, $\dot{S}_{\text{NO}}$ represents the production rate of thermal NO. The second coefficient, $\tau_{d\text{-NO}}$, is the time scale of NO destruction. The coefficients $\dot{S}_{\text{NO}}$ and $\tau_{d\text{-NO}}$ in equation (7.6) are determined by extended Zeldovich mechanism and the reburning reactions with the steady-state assumption for the atomic nitrogen. The mean rate, $\overline{\omega}_{\text{NO}}$, is obtained from the following formula:

$$\overline{\omega}_{\text{NO}} = \rho \overline{\dot{\omega}}_{\text{NO}} = \rho \int_0^\infty \int \omega_{\text{NO}}(f, \chi) P(f) P(\chi) df d\chi$$

(7.7)

The distribution of the mixture fraction in turbulent flames is represented by the beta pdf, while that of the scalar dissipation rate is given by the log-normal distribution.

In Fig. 7.1 the NO source terms ($S_{\text{NOX}}$) are shown as a function of mixture fraction for different stretch conditions. These profiles are obtained from counterflow diffusion flame calculations on the Tsuji burner configuration. The differential diffusion effect is included in the calculation. The figure shows that the source term is very sensitive to the scalar dissipation rate. At $\chi=0.058$ /s, the source term is negative in the fuel rich zone, indicating consumption of NO. At $\chi=0.424$ /s, the negative zone of the source term almost vanishes and small changes in the peak values have been found. With the increase of the scalar dissipation rate, the temperature increases while the mass fraction of O first increases, then remains less sensitive to the scalar dissipation rate and finally
decreases as the scalar dissipation rate approaches the extinction limit. At $\chi=0.424$/s, the decrease of temperature and the increase of mass fraction of O counter balance each other and the peak source term remains almost the same.

![Figure 7.1: Predicted source term $S_{\text{NOX}}$ of NO formation for different scalar dissipation rate: $\chi=0.058$; $\chi=0.424$; $\chi=2.135$; $\chi=10.653$; $\chi=37.901$; $\chi=77.05$ /s](image)

For further higher values of scalar dissipation rates, the decrease of temperature is more prominent and that reduces the source term rapidly with the increase of the scalar dissipation rate. At $\chi=77.05$/s, the formation of NO almost vanishes.

### 7.3 NOx modelling of H₂/N₂ flame

The flame considered here consists of 75% H₂ + 25% N₂ by volume and has a jet velocity of 42.3 m/s with Re=9300. 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. The comparison of the simulated and measured values for different profiles have been shown in figures 7.2 and 7.3.

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. 7.2. 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. The overall model predictions of H$_2$ are good with the exception of results in the near axis region where slight overpredictions found for X/D = 05 to X/D=40. At X/D = 60 and 80 experimental data shows no level of H$_2$ but the model predictions have shown very little amount of existence of the Hydrogen. In the prediction of mass fraction of O$_2$ profiles at all axial locations combustion model have shown fairly good results with slight overpredictions. The profiles of O$_2$ 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 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 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$_2$O are in good agreement at all the axial locations with the experiment but slight overpredictions found in the peak values and near axis region in some stations. The combustion model predicted the H$_2$O profile well in the far axis region from the peak value at x/D= 05 to 60. At x/D= 80 overprediction found between the centreline and r =90mm. Comparisons of predicted NO mass fractions in Fig. 7.2 indicate that flamelet based NOx 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 NOx 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.

Figure 7.3 shows the radial profiles of the mass fractions of $N_2$ and NO at six axial locations. Predicted NO results compared as $\text{NO} \times 100$ with $N_2/20$ to bring both in a single window. Predicted results for $N_2$ profiles are fairly good with slight overpredictions in the near axis region and underpredictions in the far axis region. The amount of nitrogen presence at different axial and radial locations of the flame seems to remain almost the same and thus the effect of $N_2$ level on NO formation cannot be identified from Fig. 7.3.
Figure 7.2: Predicted and measured mass fractions of NO, H2O, H2, O2 and mean temperature distributions in the H2/N2 jet flame. Predictions (lines); measurements (symbols).
Figure 7.3: Predicted and measured mass fractions of NO and N₂ in the H₂/N₂ jet flame. Predictions (lines); measurements (symbols).
Chapter 7. Modelling of NOx

7.4 NOx modelling of HM1 and HM3 flames

The NOx modelling for Sydney HM1 and HM3 flames has been described in this section. The HM1 flame consists of the fuel consisting of 50% CH₄ + 50% H₂ (by volume) and has a fuel jet velocity of \( U_j = 118 \, \text{m/s} \) with coflowing air velocity \( U_c = 40 \, \text{m/s} \). The only difference between HM1 and HM3 flame is that HM3 flame has the fuel jet velocity of \( U_j = 214 \, \text{m/s} \). The simulated results for different profiles have been shown in figures 7.4-7.7. The computed and measured species, temperature profiles and mass fractions of major and minor species have been presented at six axial cross-sections \( X = 13, 30, 45, 65, 90 \), and 120 mm which are equivalent to \( X/D = 0.26, 0.6, 0.9, 1.3, 1.8 \) and 2.4 after normalizing with the bluff body diameter 50mm.

Predicted results of the mass fractions of NO, CO, CO₂, H₂O and mean temperature distributions at different axial locations are compared with the measurements in Fig. 7.4 for HM1 flame. 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. The overall model predictions of H₂O are good at all the axial locations but slight overpredictions found in the peak values and near axis region in some stations. The combustion model predicted the H₂O profile well in the near axis region at all the axial locations. At \( X/D = 0.26 \) overpredicted double peak values found in the results obtained from the model. At \( x/D = 0.6 \) near axis results are found good but slight overprediction observed in the flattened maximum valued recirculation zone and in the far axis region. At \( X/D = 0.9 \) and 1.3 near axis predictions are well towards the maximum value and overpredictions found in the peak values and in the decaying region. For the axial stations \( X/D = 1.8 \) and 2.4 predicted results are fairly good with small amount of overpredictions in the peak values. In the prediction of mass fraction of CO₂ profiles at all axial locations, combustion model have shown fairly good results with slight overpredictions at all the axial locations. The profiles of CO₂ are better predicted by the model at all the axial locations but at \( X/D = 2.4 \). Predictions are even better in the case of the mass fractions of CO than CO₂. The significant amount of overpredictions 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 400K. The predicted temperature profiles have shown rapid decay from the maximum position similar to the experimental data. At X/D=0.26 and 0.6, the agreements between the measurements and the predictions are fairly good with significant amount of overpredictions by the model in the maximum valued recirculation zone. For all other axial locations better results observed in the near axis region with very little underpredictions. The temperature profiles are overpredicted in the peak values and in the decaying region for the last four axial measurement stations.

Comparisons of predicted NO mass fractions in the HM1 flame obtained from flamelet based NOx modelling have been shown in Fig 7.4. It has been observed that the model is able to predict the NO with 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 NOx levels. The computed and measured NO level have shown constant long flatten pattern of peak values in the whole of the recirculation zone. The maximum NO level observed in the hottest location of this flame and it decaying gradually in the downstream region with decreasing temperature. At X/d = 0.6 to 1.3 the peaks of NO found at the same radial positions of peak temperatures. The NO predictions and measurements show that the NO level is low in the rich part of the flame for X/D = 0.6 to 1.3. The NO level then reaches its peak further away from the axis near the flame front. Further downstream, at X/D = 1.8 and 2.4, the gradual decaying in the level of NO found with increasing radial distance.

Figure 7.5 shows the predicted and measured results of the mass fractions of NO, CO, CO2, H2O and mean temperature distributions at six axial locations for HM3 flame. As it has been done in the previous cases, predicted NO results compared as NO*500 with other species. The overall model predictions of H2O are fairly good at all the axial locations but slight overpredictions found in the peak values and in the decaying region. The combustion model predicted the H2O profile well in the near axis region at the first four axial locations but overpredictions have been observed at X/D = 1.8 and 2.4 compared to the available experimental data. In the prediction of mass fraction of CO2 profiles at all axial locations, combustion model have shown fairly good results with slight overpredictions at some axial locations. The profiles of CO2 are better predicted by the model at all the axial locations but at X/D= 1.8 and 2.4, where flamelet model
provided significant higher values compared to the available experimental data in the near axis region. Similar computational accuracy observed in the predictions of the mass fractions of CO and found better at the last two axial measurement stations.

The significant amount of overpredictions 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. The predicted temperature profiles have shown rapid decay from the maximum position similar to the experimental data. At X/D=0.26 to 1.3, predictions in the increasing near axis regions are well but overpredictions observed in the peak values and in the decaying region. At the last two measurement locations X/D= 1.8 and 2.4, model provided significant higher values in comparison with the available near axis region experimental data.

Comparisons of predicted NO mass fractions in the HM3 flame obtained from flamelet based NOx modelling have also been shown in Fig 7.5. It has been observed that the model is able to predict the NO with satisfactory 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 NOx levels. The computed and measured NO level have shown constant long flattened patterns of peak values in the recirculation zone which can be noticed from the first two locations at X/D=0.26 to 1.3. The maximum NO level observed in the hottest location of this flame and it decaying gradually in the downstream region with decreasing temperature and NO level almost vanished at the last two axial locations X/D = 1.8 and 2.4. The NO predictions and measurements show that the NO level is low in the rich part (near axis region) of the flame. 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.

Some more general remarks can be made on the formation of NO from predicted contour plots. Figures 7.6 and 7.7 show the predicted contours of the mean reaction rate, mean mass fraction of NO and mean temperature for the HM1 and HM3 flames respectively. The effects of residence time on the NO formation rate is clearly evident from these figures. For the HM1 flame, the stoichiometric mixture fraction lies on the outer edge of the outer vortex. The gas mixtures have more time to react inside the recirculation bubble and the mean NO formation rate is high. At the high jet momentum
of the HM3 flame, the stoichiometric mixture fraction lies in the inner vortex zone. In the HM3 flame, the outer vortex is shorter and the inner vortex loses its recirculation pattern due to jet expansion. The residence time in the inner vortex zone is much shorter than that in the outer vortex and hence the NO-formation rate is much less in the HM3 flame. The mean reaction rate contours resemble the mean temperature contours. However, NO-formation is only confined to the high temperature region along the flame front. For both the flames, the mean NO-formation rate is low in the neck zone and increases further downstream. Though the mean NO-formation is confined along the stoichiometric mixture fraction, the contour plots show that the mean NO exists in a wide area of the flames due to the convection and diffusion processes.
Figure 7.4: Computed and measured mass fractions of NO, CO, CO$_2$, H$_2$O and mean temperature distributions in the HM1 flame. Computation (lines); measurements (symbols) [Sydney data].
Figure 7.5: Computed and measured mass fractions of NO, CO, CO$_2$, H$_2$O and mean temperature distributions in the HM3 flame. Computations (lines); measurements (symbols) [Sydney data].
Figure 7.6: Predicted contours of (a) mean reaction rates $\dot{\omega}_{NO}$, (b) mean mass fractions of NO and (c) temperatures in the HM1 flame
Figure 7.7: Predicted contours of (a) mean reaction rates $\dot{\omega}_{\text{NO}}$, (b) mean mass fractions of NO and (c) temperatures in the HM3 flame.
Chapter 7. Modelling of NOx

7.5 NOx Reductions

Although NOx reduction computation has not been carried out in this study but a brief description of the available NO reduction methods have been made in this section. There are two main different methods of reduction of NO are in picture at present, which are the NO reduction by combustion modification, NO reduction by post-combustion processes like catalytic combustion.

7.5.1 NO reduction by combustion modification

Combustion modifications are often called primary measures. It is hoped that combustion modifications are without great cost and that they do not need any addition of other compounds. On the other hand, primary methods typically have special new geometrical requirements with the combustion device, and it is very difficult to change old combustion devices to meet those requirements. Thus primary methods are normally used in new combustion devices and for old combustion devices there are the possibilities to use secondary methods, which will be described in the next section.

Because of the high activation energy \( T_a \approx 38200 \, \text{K} \) of thermal NO mechanism any scheme that suppresses peak temperatures will lower the NO output. Thus the \( \text{N}_2 \), which is formed in the first stage, is not converted to thermal NO, because the combustion temperature is steadily reduced due to radiative and convective heat transfer. In nonpremixed jet flames, the radiation from the flame, which lowers peak temperature, has a significant effect on NO generated. It would be beneficial to inject an inert diluent gas such as nitrogen or water, whose additional heat capacity lowers the peak temperature. For this purpose, exhaust gases are reasonably inert. When this effective process is done in piston engines, it is called exhaust-gas recirculation (EGR), and when done in atmospheric boiler flame, flue-gas recirculation (FGR). Even though the success of EGR, the high temperatures and pressures inside the Diesel and Otto engines promotes NO formation. For this reason, devices that burn at lower temperature and pressure are receiving increased attention.

NOx formation in gas turbines is reviewed by Correa (1992). Water injection steadily lowers NOx output until the mass flow rate of water is about equal to the fuel mass flow
rate, at which point the amount of CO and unburnt hydrocarbons increases rapidly to unacceptable levels. Of course in any combustion system, the longer the high temperature residence time, the closer to equilibrium NO the system becomes. Thus an optimum time is desired such that all of the fuel is oxidized, as well as intermediates such as CO, and the formation of NO is terminated by rapid cooling (Takeno et al. 1993)

7.5.2 NO reduction by catalytic combustion

Post combustion processes (or secondary measures) are necessary to remove pollutants like NO when combustion modifications are not efficient enough or not possible at all. Most well known NO reduction method is the catalytic converter that is in the exhaust system of many automobiles (Heywood 1988). The catalyst is a remarkable combination of noble metals that oxidize CO to CO$_2$ and simultaneously reduce NO to N$_2$. Key success of the catalytic converter is the $\lambda$-sensor that detects if there is any O$_2$ in the exhaust. If O$_2$ is detected, electronic feedback control to the engine directs a slow increase in the fuel flow-rate, and when no O$_2$ is detected, the fuel flow-rate is slowly decreased. Thus on average the engine operates at stoichiometric conditions ($\Phi = \lambda = 1$) and likewise, catalyst operates in a low O$_2$ and low fuel environment.

Power plants like furnaces, gas turbines and some Diesel engines, often used catalyst with addition of ammonia which NO on the catalyst to produce N$_2$ and water. The selective catalytic reduction (SCR) catalyst is active over a wide range of temperatures but is sensitive to fouling from particulates and sulphur in the exhaust (Bowman 1993). Further NO reduction and increase in thermal efficiency may be achieved by reforming the fuel first via global reactions like CH$_4$ + H$_2$O = CO + 3H$_2$ which is endothermic. Combustion of reformed fuel gives even lower NO due to lack of CH and the leaner flammability limit of H$_2$- containing fuels.
7.6 Discussion

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, Syney HM1 and HM3 flames. 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 compositions 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 results presented here are obtained from the flamelet model with differential diffusion effects, as the formation of NO is found quite sensitive to differential diffusion effects. As a test case, an improved prediction by the ad-hoc variable Lewis number flamelet calculation implies that the correct handling of the differential diffusion effects is very important to improve the agreement. However, the correct handling of the extent of the differential diffusion effects in the turbulent flames is lacking in the present modelling concept and it is essential to develop a model to include this effect.

The NO formed through the thermal route has been considered here. As described above and Chen and Chang (1996) have also shown that the Zeldovich mechanism is the dominant pathways for the production of NO in a turbulent jet flames except in the rich parts of the flame. In the rich part of the flame, they have shown that the NO is consumed mainly through the reaction step HCCO+NO=CO+HCNO. The importance of NO reburning through the HCN and CN intermediates in the rich system is described by a number of researchers (Miller and Bowman 1989; Dupont et al. 1995). Thus, the overprediction of NO in the fuel rich side in some cases and in the fuel lean decaying region can be attributed to the non-inclusion of the NO reburn mechanism.
7.7 Summary

1. Four different routes of formation and emission of NO\(_x\) are described in brief.

2. The performance of the flamelet based NO modelling technique is appraised against the single point measurements in the H\(_2\)/N\(_2\) jet diffusion and Sydney HM1 and HM3 bluff body stabilised flames.

3. The flamelet based model yielded reasonable predictions of NO with the thermal NO mechanism. The predicted results of temperature and major and minor species as well as NO are compared against the reported experimental data. The predictions in the peak values of temperatures and some species are somehow not well agreed with data.

4. Although the emission of NO is adequately represented by the Zeldovich mechanism, it has been shown that the prediction of NO in the fuel rich side in some cases and in the fuel lean decaying region is not adequate but the NO reburn mechanism may produce better results.

5. Temperature and concentrations of major and minor species other than NO are obtained by appropriately averaging the flamelets, whereas the concentration of NO is calculated by solving its own transport equation with the source term obtained from the flamelet library.

6. 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.

7. Finally, a brief description of the main available NO\(_x\) reduction methods has been made.
Concluding Remarks

In the previous chapters, various aspects of the turbulent combustion modelling have been investigated and discussed. This concluding chapter is devoted to the discussion of the main conclusions of this study. The conclusions of the main focal points of this study, the assessment of combustion models, combustion radiation modelling and flamelet based NOx modelling strategies have been drawn in the following sections.

To assess different combustion models and to use the appropriate model in simulation an in-house CFD code has been developed. The flame sheet, equilibrium and laminar flamelet models with unity Lewis number and differential diffusion effects are incorporated in the CFD code. The influence of the numerical error associated with the discretisation scheme is assessed by comparing the results obtained by the hybrid, power law and TVD schemes. Flames were simulated using the computer code RUN-1DL, developed by Rogg (1995). This algorithm allows for mixture-averaged multicomponent diffusion, thermal diffusion, variable thermochemical properties, and variable transport properties. The CHEMKIN II package of Sandia national laboratories Kee et al. (1993) was used as a preprocessor to find the thermochemical and transport properties for RUN-1DL.

The configurations used for the validation and assessment of the combustion models are co-flowing jet flames and bluff body burners. The high quality experimental databases available from Sandia national laboratories, the University of Sydney and other reported measurements are used for the purpose of evaluating the combustion models. The reasons behind these burners selections are that the jet flames have many practical applications and bluff-body burner provides a flame suitable for the study of turbulence-chemistry interactions with a great similarity to practical combustors used in many industrial applications. The bluff-body burner is, therefore, a suitable compromise
as a model problem because it has some of the complications associated with practical combustors while preserving relatively simple and well-defined boundary conditions. The selected jet flames also provides a controlled environment for the study of turbulence-chemistry interaction. The combustion of different fuels ranging from simple $\text{H}_2/\text{N}_2$ and $\text{CO}/\text{H}_2/\text{N}_2$ to complex $\text{CH}_4/\text{H}_2$ are investigated for different inlet velocities and boundary conditions. The predicted results demonstrate the effects of turbulent mixing and the effects of chemical reactions on the combustion models.

8.1 Assessments of combustion models

The combustion models are assessed by comparing radial profiles of mean mixture fraction, variance of mixture fraction, temperature and concentrations of major and minor species at different axial locations. The performances of the flame sheet, chemical equilibrium and laminar flamelet models are analysed for predicting temperature, major and minor species for a number of jet diffusion flames. In flamelet model unity Lewis number and the effects of differential diffusion are considered. The comparison of the predictions and the measurements in the HN1, HN2 and HN3 flames obtained from $\text{H}_2/\text{N}_2$ fuels of different inlet velocities and compositions provides insight into the behaviour of the near equilibrium chemistry regime.

The results presented demonstrate that the flamelet models predictions based on the differential diffusion as well as with unity Lewis number are, in general, both qualitatively and quantitatively superior to those of other models. Overall predictions of major and minor species and flame temperatures, in all three flames are in reasonable agreement with experimental data. Results do, however, tend to unpredict mixture fractions, temperatures and species mass fractions in some cases. The effect of the differential diffusion was studied and found to be less important for the prediction of $\text{H}_2/\text{N}_2$ flame but obviously it is more significant in the cases of some other flames. The unity Lewis number flamelet has provided a better representation of transport of the species. The influence of different non-equilibrium parameters was evaluated and only the scalar dissipation rate was found to be suitable as a non-equilibrium parameter. The modified $k$-$\varepsilon$ model with $C_{e1} = 1.60$ provides more accurate results and couplings of chemical models with turbulence were found well suited for the $\text{H}_2/\text{N}_2$ flames. The increase of jet momentum results in longer flames observed from the stoichiometric
mixture fraction contours. The differences in the predicted results for different discretisation schemes were found to be very small.

The turbulence intensity along the flame front is relatively low, which is expected to be dominated by the large structures. The reaction zones are stretched by the large scales of turbulence, which will not interfere with the inner structure of flames. Large structures also quench the flame locally. In the flamelet model, local extinction is predicted when the scalar dissipation rate reaches a quenching limit and the blow-off of the flame occurs when the proportion of burning flamelets decreases below a certain limit. However, the flamelet model in the present study shows that this simple treatment is not adequate to predict the local extinction and subsequent lower temperatures in the jet flames. The present study concludes that the laminar flamelet model is capable of predicting flames which are far away from the equilibrium condition, provided the localised extinction is absent in the flames.

Based on the performance of the combustion models in a number of flames with different types of fuel and turbulent mixing rate, a number of recommendations may be made for turbulent flame calculations. The flame-sheet, eddy break-up and equilibrium models are found to be inadequate even for the near equilibrium flames. Therefore, more advanced models have to be used to provide a better description of the flame. In situations, where finite rate chemistry effects are important the laminar flamelet model is a good choice. In this context, it is important to distinguish between two types of finite-rate chemistry effects. The first type of the finite-rate chemistry effect that are caused by the interaction of the turbulent mixing and chemical kinetics, but which do not lead to local extinction. The laminar flamelet model is capable of predicting this type of finite-rate chemistry effect. In the second type, where the turbulent mixing leads to considerable local extinction even though the mean stretching rate is below the extinction limit. The laminar flamelet model is not capable of predicting the flames where local extinction is observed. The transient effects are very important when localised extinction occurs. The prediction of localised extinction requires a detailed analysis of the time scales involving reaction, convection and diffusion processes and there is future possibility that with a transient flamelet model, prediction problem of the localised extinction phenomenon may be resolved.
8.2 Combustion radiation modelling

The flame radiation plays an important role and being the dominant mode of heat transfer in fires and in many high temperature combustion systems. The flamelet modelling of diffusion flames and coupling between the radiation heat transfer and flamelet model has been studied. The effect of radiation heat transfer on the predictive capability of the flamelet combustion model is investigated. The coupling between the radiation and combustion is achieved through an additional parameter, the so-called enthalpy defect, which is defined as the difference between the actual enthalpy and the adiabatic enthalpy of a flame. The flamelet library with the required enthalpy defect is generated by modifying the available enthalpy at the boundaries.

The overall computational strategy combines the modelling of finite rate chemistry effects with the detailed solution of the accompanying coupled equation of radiative exchange. The radiative heat exchange is computed by the discrete transfer model and the gas radiation properties are established from the concentrations of major and minor species available from the combustion calculations. A weighted sum of gray gases (WSGG) method is employed for the radiation properties calculations. Radiation properties, emissivities and intensities have been computed from the mean values of the scalar variables. The effect of the radiation heat transfer on temperature and mass fractions of major species and minor species like OH and NO are studied and comprehensive validation of CFD based flamelet/radiation results for reacting flows is shown that for the geometries presented here, the effect of radiation is important at regions high temperatures where the temperatures are around 2000 K. The effects of the radiation are found very important for OH and NO predictions. The inclusion of radiation heat transfer into the simulation remarkably improves the prediction of OH mass fraction. Thermal radiation from flames reduces the local temperatures sufficiently to affect the production rate of NO and the incorporation of radiation heat transfer in combustion model is found to improve the prediction of pollutant emissions such as NO species significantly.

The present study demonstrates that the radiation adjusted flamelet model for combustion leads to encouraging results that reproduce the overall characteristics of
radiating turbulent diffusion flames and the flamelet/radiation modelling strategy presented here has potential for further improvement.

8.3 Flamelet modelling of NOx

The performance of the flamelet based NO modelling technique is appraised against the single point measurements of diffusion flames. The flamelet combustion model provides an efficient method to model the H₂/N₂ jet diffusion flame and Sydney HM1 and HM3 flames. 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 compositions 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 results presented here are obtained from the flamelet model with differential diffusion effects, as the formation of NO is found quite sensitive to differential diffusion effects. The detailed analysis of the predicted results under different fuels and flow conditions has indicated that the laminar flamelet model is the best choice for predicting temperature and major and minor species concentrations like OH and NO.

The current understanding of differential diffusion effect is still very limited. From the modelling point of view, it is in principle, possible to account for the differential diffusion effects. For this to happen, a large quantity of experimental data is required to identify and correlate the differential diffusion parameter with the Reynolds number, chemical reaction e.g. the stoichiometric mixture fraction and the reaction zone thickness, and types of parent fuel. Based on the observed correlation, the model for the differential diffusion can be developed, which in turn can be utilised in the flamelet method of turbulent combustion prediction.

Although the emission of NO is adequately represented by the Zeldovich mechanism, it has been shown that the prediction of NO in the fuel rich side in some cases and in the fuel lean decaying region is not adequate but the NO reburn mechanism may produce better results. 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.

The present study has also shown that the laminar flamelet model is well capable of predicting temperature and concentrations of major and minor species for flames at near equilibrium as well as away from equilibrium but without the local extinction. The effects of differential diffusion is crucial for prediction of the dynamics of the near-field, where chemical kinetics and transport of radicals, which have a broad range of diffusivities, are important. It has been shown that only the scalar dissipation rate as the non-equilibrium parameter can reproduce the correct trend of the flame structure and the unity Lewis number flamelet gives a better representation of the transport of the species in turbulent flames.

8.4 Recommendations for future work

Many novel issues have been raised from the present study; future broad ranges of research possibilities and extended ideas of the present research are identified. Some of those are:

- The laminar flamelet model should be modified to include the actual differential diffusion effects instead of the current approach of either setting full molecular transport or setting the equal mass diffusivities and unity Lewis number.

- The flamelet radiation combustion model has a lot of potential for further development. The advanced property calculation algorithms should be utilised and effects of turbulence/radiation interactions should be investigated.

- The transient effects in the laminar flamelet modelling should be investigated, specially for the prediction of CO and local extinction in hydrocarbon flames.

- The sensitivity of the different reaction mechanisms on the prediction capability of the laminar flamelet model should be studied and different pathways of NO formation should be implemented in the laminar flamelet 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 with differential diffusion effects 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.

Flamelet modelling with the incorporation of LES (large eddy simulation) turbulence model, which may be regarded as Flamelet-LES modelling approach.
Appendix A

Table A.1: Detailed Chemical Mechanisms and their rate parameters in units of mol, cm, s, kJ and K. Reference: Peters (1993)

<table>
<thead>
<tr>
<th>No.</th>
<th>Reactions</th>
<th>$B_\beta$</th>
<th>$a_\beta$</th>
<th>$E_\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td><strong>H$_2$/O$_2$ Chain Reactions</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1f</td>
<td>$\text{O}_2 + \text{H} \rightarrow \text{OH} + \text{O}$</td>
<td>$2.000\times10^{14}$</td>
<td>0.00</td>
<td>70.30</td>
</tr>
<tr>
<td>1b</td>
<td>$\text{OH} + \text{O} \rightarrow \text{O}_2 + \text{H}$</td>
<td>$1.568\times10^{13}$</td>
<td>0.00</td>
<td>3.52</td>
</tr>
<tr>
<td>2f</td>
<td>$\text{H}_2 + \text{O} \rightarrow \text{OH} + \text{H}$</td>
<td>$5.060\times10^{04}$</td>
<td>2.67</td>
<td>26.30</td>
</tr>
<tr>
<td>2b</td>
<td>$\text{OH} + \text{H} \rightarrow \text{H}_2 + \text{O}$</td>
<td>$2.222\times10^{04}$</td>
<td>2.67</td>
<td>18.29</td>
</tr>
<tr>
<td>3f</td>
<td>$\text{H}_2 + \text{OH} \rightarrow \text{H}_2\text{O} + \text{H}$</td>
<td>$1.000\times10^{08}$</td>
<td>1.06</td>
<td>13.80</td>
</tr>
<tr>
<td>3b</td>
<td>$\text{H}_2\text{O} + \text{H} \rightarrow \text{H}_2 + \text{OH}$</td>
<td>$4.312\times10^{08}$</td>
<td>1.06</td>
<td>76.46</td>
</tr>
<tr>
<td>4f</td>
<td>$\text{OH} + \text{OH} \rightarrow \text{H}_2\text{O} + \text{O}$</td>
<td>$1.500\times10^{09}$</td>
<td>1.14</td>
<td>0.42</td>
</tr>
<tr>
<td>4b</td>
<td>$\text{H}_2\text{O} + \text{O} \rightarrow \text{OH} + \text{OH}$</td>
<td>$1.473\times10^{10}$</td>
<td>1.14</td>
<td>71.09</td>
</tr>
<tr>
<td></td>
<td><strong>HO$_2$ formation and Consumption</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5f</td>
<td>$\text{O}_2 + \text{H} + \text{M}' \rightarrow \text{HO}_2 + \text{M}'$</td>
<td>$2.300\times10^{18}$</td>
<td>-0.80</td>
<td>0.00</td>
</tr>
<tr>
<td>5b</td>
<td>$\text{HO}_2 + \text{M}' \rightarrow \text{O}_2 + \text{H} + \text{M}'$</td>
<td>$3.190\times10^{18}$</td>
<td>-0.80</td>
<td>195.39</td>
</tr>
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<td>6</td>
<td>$\text{HO}_2 + \text{H} \rightarrow \text{OH} + \text{OH}$</td>
<td>$1.500\times10^{14}$</td>
<td>0.00</td>
<td>4.20</td>
</tr>
<tr>
<td>7</td>
<td>$\text{HO}_2 + \text{H} \rightarrow \text{H}_2 + \text{O}_2$</td>
<td>$2.500\times10^{13}$</td>
<td>0.00</td>
<td>2.90</td>
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<tr>
<td>8</td>
<td>$\text{HO}_2 + \text{OH} \rightarrow \text{H}_2\text{O} + \text{O}_2$</td>
<td>$6.000\times10^{13}$</td>
<td>0.00</td>
<td>0.00</td>
</tr>
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<td>9</td>
<td>$\text{HO}_2 + \text{H} \rightarrow \text{H}_2\text{O} + \text{O}$</td>
<td>$3.000\times10^{13}$</td>
<td>0.00</td>
<td>7.20</td>
</tr>
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<td>10</td>
<td>$\text{HO}_2 + \text{O} \rightarrow \text{OH} + \text{O}_2$</td>
<td>$1.800\times10^{13}$</td>
<td>0.00</td>
<td>-1.70</td>
</tr>
<tr>
<td></td>
<td><strong>H$_2$O$_2$ formation and Consumption</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>11</td>
<td>$\text{HO}_2 + \text{HO}_2 \rightarrow \text{H}_2\text{O}_2 + \text{O}_2$</td>
<td>$2.500\times10^{11}$</td>
<td>0.00</td>
<td>-5.20</td>
</tr>
<tr>
<td>12f</td>
<td>$\text{OH} + \text{OH} + \text{M}' \rightarrow \text{H}_2\text{O}_2 + \text{M}'$</td>
<td>$3.250\times10^{22}$</td>
<td>-2.00</td>
<td>0.00</td>
</tr>
<tr>
<td>12b</td>
<td>$\text{H}_2\text{O}_2 + \text{M}' \rightarrow \text{OH} + \text{OH} + \text{M}'$</td>
<td>$1.692\times10^{24}$</td>
<td>-2.00</td>
<td>202.29</td>
</tr>
</tbody>
</table>

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<table>
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<tr>
<th>No.</th>
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<th>$a_\beta$</th>
<th>$E_\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>13</td>
<td>$H_2O_2 + H \rightarrow H_2O + OH$</td>
<td>1.000E+13</td>
<td>0.00</td>
<td>15.00</td>
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<tr>
<td>14f</td>
<td>$H_2O_2 + H \rightarrow H_2 + HO_2$</td>
<td>1.700E+12</td>
<td>0.00</td>
<td>15.70</td>
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<td>14b</td>
<td>$H_2 + HO_2 \rightarrow H_2O_2 + H$</td>
<td>1.150E+12</td>
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**Recombination Reactions**

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<tr>
<td>15</td>
<td>$H + H + M' \rightarrow H_2 + M'$</td>
<td>1.800E+18</td>
<td>-1.00</td>
<td>0.00</td>
</tr>
<tr>
<td>16</td>
<td>$OH + H + M' \rightarrow H_2O + M'$</td>
<td>2.200E+22</td>
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<td>0.00</td>
</tr>
<tr>
<td>17</td>
<td>$O + O + M' \rightarrow O_2 + M'$</td>
<td>2.900E+17</td>
<td>-1.00</td>
<td>0.00</td>
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**CO/CO₂ Mechanism**

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<th>$E_\beta$</th>
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</thead>
<tbody>
<tr>
<td>18f</td>
<td>$CO + OH \rightarrow CO_2 + H$</td>
<td>4.400E+06</td>
<td>1.50</td>
<td>-3.10</td>
</tr>
<tr>
<td>18b</td>
<td>$CO_2 + H \rightarrow CO + OH$</td>
<td>4.956E+08</td>
<td>1.50</td>
<td>89.76</td>
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**CH Consumption**

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<th>$E_\beta$</th>
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</thead>
<tbody>
<tr>
<td>19</td>
<td>$CH + O_2 \rightarrow CHO + O$</td>
<td>3.000E+13</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>20</td>
<td>$CO_2 + CH \rightarrow CHO + CO$</td>
<td>3.400E+12</td>
<td>0.00</td>
<td>2.90</td>
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**CHO Consumption**

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<th>$E_\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>21</td>
<td>$CHO + H \rightarrow CO + H_2$</td>
<td>2.000E+14</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>22</td>
<td>$CHO + OH \rightarrow CO + H_2O$</td>
<td>1.000E+14</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>23</td>
<td>$CHO + O_2 \rightarrow CO + HO_2$</td>
<td>3.000E+12</td>
<td>0.00</td>
<td>0.00</td>
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<tr>
<td>24f</td>
<td>$CHO + M' \rightarrow CO + H+ M'$</td>
<td>7.100E+14</td>
<td>0.00</td>
<td>70.30</td>
</tr>
<tr>
<td>24b</td>
<td>$CO + H+ M' \rightarrow CHO + M'$</td>
<td>1.136E+15</td>
<td>0.00</td>
<td>9.97</td>
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**CH₂ Consumption**

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<tr>
<td>25f</td>
<td>$CH_2 + H \rightarrow CH + H_2$</td>
<td>8.400E+09</td>
<td>1.50</td>
<td>1.40</td>
</tr>
<tr>
<td>25b</td>
<td>$CH + H_2 \rightarrow CH_2 + H$</td>
<td>5.830E+09</td>
<td>1.50</td>
<td>13.08</td>
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<td>26</td>
<td>$CH_2 + O \rightarrow CO+H + H$</td>
<td>8.000E+13</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>27</td>
<td>$CH_2 + O_2 \rightarrow CO + OH + H$</td>
<td>6.500E+12</td>
<td>0.00</td>
<td>6.30</td>
</tr>
<tr>
<td>28</td>
<td>$CH_2 + O_2 \rightarrow CO_2 + H + H$</td>
<td>6.500E+12</td>
<td>0.00</td>
<td>6.30</td>
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**CH₂O Consumption**

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</thead>
<tbody>
<tr>
<td>29</td>
<td>$CH_2O + H \rightarrow CHO + H_2$</td>
<td>2.500E+13</td>
<td>0.00</td>
<td>16.70</td>
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<table>
<thead>
<tr>
<th>No.</th>
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<th>$E_\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>30</td>
<td>CH$_2$O + O $\rightarrow$ CHO + OH</td>
<td>3.500E+13</td>
<td>0.00</td>
<td>14.60</td>
</tr>
<tr>
<td>31</td>
<td>CH$_2$O + OH $\rightarrow$ CHO + H$_2$O</td>
<td>3.000E+13</td>
<td>0.00</td>
<td>5.00</td>
</tr>
<tr>
<td>32</td>
<td>CH$_2$O + $M'$ $\rightarrow$ CHO + H + $M'$</td>
<td>1.400E+17</td>
<td>0.00</td>
<td>320.00</td>
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<tr>
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<td>CH$_3$ Consumption</td>
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<tr>
<td>33f</td>
<td>CH$_3$ + H $\rightarrow$ CH$_2$ + H$_2$</td>
<td>1.800E+14</td>
<td>0.00</td>
<td>63.00</td>
</tr>
<tr>
<td>33b</td>
<td>CH$_2$ + H$_2$ $\rightarrow$ CH$_3$ + H</td>
<td>3.680E+13</td>
<td>0.00</td>
<td>44.30</td>
</tr>
<tr>
<td>34</td>
<td>CH$_3$ + H + (M)$^1$ $\rightarrow$ CH$_4$ + (M)</td>
<td>$k_\infty$</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$k_0$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>35</td>
<td>CH$_3$ + O$_2$ $\rightarrow$ CH$_2$O + OH</td>
<td>7.000E+13</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>36</td>
<td>CH$_3$ + CH$_3$ + (M) $\rightarrow$ C$_2$H$_6$ + (M)</td>
<td>$k_\infty$</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$k_0$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>37</td>
<td>CH$_3$ + O$_2$ $\rightarrow$ CH$_2$O + OH</td>
<td>3.400E+11</td>
<td>0.00</td>
<td>37.40</td>
</tr>
<tr>
<td>38f</td>
<td>CH$_4$ + H $\rightarrow$ CH$_3$ + H$_2$</td>
<td>3.400E+04</td>
<td>3.00</td>
<td>36.60</td>
</tr>
<tr>
<td>38b</td>
<td>CH$_3$ + H$_2$ $\rightarrow$ CH$_4$ + H</td>
<td>3.400E+02</td>
<td>3.00</td>
<td>34.56</td>
</tr>
<tr>
<td>39</td>
<td>CH$_4$ + O $\rightarrow$ CH$_3$ + OH</td>
<td>3.400E+07</td>
<td>2.10</td>
<td>31.90</td>
</tr>
<tr>
<td>40f</td>
<td>CH$_4$ + OH $\rightarrow$ CH$_3$ + H$_2$O</td>
<td>3.400E+06</td>
<td>2.10</td>
<td>10.30</td>
</tr>
<tr>
<td>40b</td>
<td>CH$_3$ + H$_2$O $\rightarrow$ CH$_4$ + OH</td>
<td>3.400E+05</td>
<td>2.10</td>
<td>70.92</td>
</tr>
</tbody>
</table>

Third body efficiencies with respect to molecular hydrogen are: 6.5 for CH$_4$, 6.5 for H$_2$O, 1.5 for CO$_2$, 1.0 for H$_2$, 0.75 for CO, 0.4 for O$_2$, 0.4 N$_2$ and 1.0 for all other species.

$^1$Lindemann form, $k = k_\infty / (1 + k_{f\text{ all}} / [M])$, where $k_{f\text{ all}} = 0.0063 \exp(-18000/ RT)$ and the molecular density of the third body is the sum of molar densities of all species, weighted with their catalytic efficiency $z_i$, $[M] = \sum_{i=1}^{n} z_i [X_i]$. 
Appendix B

Table B.1: Reaction Mechanisms of NO\textsubscript{x} formation and their rate parameters in units of mol, cm, s, kJ and K. (Klaus and Warnatz 1995)

<table>
<thead>
<tr>
<th>No.</th>
<th>Reactions</th>
<th>$B_\beta$</th>
<th>$a_\beta$</th>
<th>$E_\beta$</th>
</tr>
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<tbody>
<tr>
<td>30 - 40: Reactions of H-N-O Species</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td><strong>Consumption of NH\textsubscript{3}</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>30</td>
<td>$\text{NH}_3 + \text{H} = \text{NH}_2 + \text{H}_2$</td>
<td>6.36E+05</td>
<td>2.40</td>
<td>42.60</td>
</tr>
<tr>
<td></td>
<td>$\text{NH}_3 + \text{O} = \text{NH}_2 + \text{OH}$</td>
<td>1.10E+06</td>
<td>2.10</td>
<td>21.80</td>
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<tr>
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<td>$\text{NH}_3 + \text{OH} = \text{NH}_2 + \text{H}_2\text{O}$</td>
<td>2.04E+06</td>
<td>2.00</td>
<td>2.37</td>
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<tr>
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<td>$\text{NH}_3 + \text{M}^* = \text{NH}_2 + \text{H} + \text{M}^*$</td>
<td>1.40E+16</td>
<td>0.06</td>
<td>379.0</td>
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<tr>
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<td><strong>Consumption of NH\textsubscript{2}</strong></td>
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<td></td>
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<tr>
<td>31</td>
<td>$\text{NH}_2 + \text{H} = \text{NH} + \text{H}_2$</td>
<td>6.00E+12</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
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<td>$\text{NH}_2 + \text{O} = \text{NH} + \text{OH}$</td>
<td>7.00E+12</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td>$\text{NH}_2 + \text{O} = \text{HNO} + \text{OH}$</td>
<td>4.50E+13</td>
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<td>0.00</td>
</tr>
<tr>
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<td>$\text{NH}_2 + \text{O} = \text{NO} + \text{H}_2$</td>
<td>5.00E+12</td>
<td>0.00</td>
<td>0.00</td>
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<td>$\text{NH}_2 + \text{N} = \text{N}_2 + \text{H} + \text{H}$</td>
<td>7.20E+13</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td>$\text{NH}_2 + \text{O}_2 = \text{HNO} + \text{OH}$</td>
<td>4.50E+12</td>
<td>0.00</td>
<td>105.0</td>
</tr>
<tr>
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<td>$\text{NH}_2 + \text{O}_2 = \text{NH} + \text{HO}_2$</td>
<td>1.00E+14</td>
<td>0.00</td>
<td>209.0</td>
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<tr>
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<td>$\text{NH}_2 + \text{OH} = \text{NH} + \text{H}_2\text{O}$</td>
<td>9.00E+07</td>
<td>1.50</td>
<td>-1.91</td>
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<tr>
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<td>$\text{NH}_2 + \text{HO}_2 = \text{NH}_3 + \text{O}_2$</td>
<td>4.50E+13</td>
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<td>0.00</td>
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<td>$\text{NH}_2 + \text{NH}_2 = \text{NH}_3 + \text{NH}$</td>
<td>6.30E+12</td>
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<td>41.8</td>
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<td><strong>Consumption of NH</strong></td>
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</tr>
<tr>
<td>32</td>
<td>$\text{NH} + \text{H} = \text{N} + \text{H}_2$</td>
<td>1.00E+13</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td>$\text{NH} + \text{O} = \text{NO} + \text{H}$</td>
<td>7.00E+13</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td>$\text{NH} + \text{OH} = \text{NO} + \text{H}_2$</td>
<td>2.40E+13</td>
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<td>0.00</td>
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<tr>
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<td>$\text{NH} + \text{OH} = \text{NO} + \text{H}_2$</td>
<td>2.00E+09</td>
<td>1.20</td>
<td>0.02</td>
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<td>$\text{NH} + \text{OH} = \text{NO} + \text{H}_2$</td>
<td>4.00E+13</td>
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<td>0.00</td>
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<tr>
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<th>$\alpha_\beta$</th>
<th>$E_\beta$</th>
</tr>
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<tr>
<td></td>
<td>$\text{NH}^+ O_2 = \text{NO} + \text{OH}$</td>
<td>1.00E+13</td>
<td>-0.20</td>
<td>20.80</td>
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<tr>
<td></td>
<td>$\text{NH}^+ O_2 = \text{HNO} + O$</td>
<td>4.60E+05</td>
<td>2.00</td>
<td>27.20</td>
</tr>
<tr>
<td></td>
<td>$\text{NH}^+ \text{NH} = \text{N}_2 + \text{H} + \text{H}$</td>
<td>2.54E+13</td>
<td>0.00</td>
<td>0.40</td>
</tr>
</tbody>
</table>

**Consumption of N**

|     | $\text{N} + \text{OH} = \text{NO} + \text{H}$ | 3.80E+13  | 0.00           | 0.00      |
|     | $\text{N} + O_2 = \text{NO} + O$              | 6.40E+09  | 1.00           | 26.1      |
|     | $\text{N} + \text{CO}_2 = \text{NO} + \text{CO}$ | 1.90E+11  | 0.00           | 14.2      |
|     | $\text{N} + \text{NO} = \text{N}_2 + O$      | 3.27E+12  | 0.30           | 0.00      |
|     | $\text{N} + \text{N} + \text{M}^* = \text{N}_2 + \text{M}^*$ | 2.26E+17  | 0.00           | 32.3      |
|     | $\text{N} + \text{NH} = \text{N}_2 + \text{H}$ | 3.00E+13  | 0.00           | 0.00      |
|     | $\text{N} + \text{CH} = \text{CN} + \text{H}$ | 1.30E+12  | 0.00           | 0.00      |
|     | $\text{N} + 3\text{CH}_2 = \text{HCN} + \text{H}$ | 5.00E+13  | 0.00           | 0.00      |
|     | $\text{N} + \text{CH}_3 = \text{H}_2\text{CN} + \text{H}$ | 7.10E+13  | 0.00           | 0.00      |
|     | $\text{N} + \text{HCCO} = \text{HCN} + \text{CO}$ | 5.00E+13  | 0.00           | 0.00      |
|     | $\text{N} + \text{C}_2\text{H}_2 = \text{HCN} + \text{CH}$ | 1.04E+13  | -0.50          | 0.00      |
|     | $\text{N} + \text{C}_2\text{H}_3 = \text{HCN} + 3\text{CH}_2$ | 2.00E+13  | 0.00           | 0.00      |

**Consumption of N$_2$H**

|     | $\text{N}_2\text{H} + \text{O} = \text{N}_2\text{O} + \text{H}$ | 1.00E+14  | 0.00           | 0.00      |
|     | $\text{N}_2\text{H} + \text{O} = \text{NO} + \text{NH}$         | 1.00E+13  | 0.00           | 0.00      |
|     | $\text{N}_2\text{H} + \text{OH} = \text{N}_2 + \text{H}_2\text{O}$ | 3.00E+13  | 0.00           | 0.00      |
|     | $\text{N}_2\text{H} + \text{M}^* = \text{N}_2 + \text{H} + \text{M}^*$ | 1.70E+12  | 0.00           | 59.9      |
|     | $\text{N}_2\text{H} + \text{NO} = \text{N}_2 + \text{HNO}$       | 5.00E+13  | 0.00           | 0.00      |

**Consumption of N$_2$**

|     | $\text{N}_2 + \text{CH} = \text{HCN} + \text{N}$               | 1.56E+11  | 0.00           | 75.10     |
|     | $\text{N}_2 + 3\text{CH}_2 = \text{HCN} + \text{NH}$           | 4.28E+12  | 0.00           | 150.0     |

**Consumption of NO**

|     | $\text{NO} + \text{OH} + \text{M}^* = \text{HNO}_2 + \text{M}^*$ | 5.08E+12  | -2.50          | 0.28      |
|     | $\text{NO} + \text{HO}_2 = \text{NO}_2 + \text{OH}$             | 2.10E+12  | 0.00           | -2.01     |
|     | $\text{NO} + \text{NH} = \text{N}_2 + \text{OH}$                 | 2.16E+13  | -0.23          | 0.00      |

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<table>
<thead>
<tr>
<th>No.</th>
<th>Reactions</th>
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<th>$a_{\beta}$</th>
<th>$E_{\beta}$</th>
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<tbody>
<tr>
<td></td>
<td>NO + NH  = N$_2$O + H</td>
<td>2.94E+14</td>
<td>-0.40</td>
<td>0.00</td>
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<tr>
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<td>NO + NH  = N$_2$O + H</td>
<td>-2.16E+13</td>
<td>-0.23</td>
<td>0.00</td>
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<tr>
<td></td>
<td>NO + NH$_2$ = N$_2$ + H$_2$O</td>
<td>2.00E+20</td>
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<td>3.87</td>
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<td>NO + NH$_2$ = N$_2$ + H + OH</td>
<td>4.76E+15</td>
<td>-1.10</td>
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<tr>
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<td>NO + NH$_2$ = N$_2$H + OH</td>
<td>3.97E+11</td>
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<td>-1.63</td>
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<tr>
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<td>NO + CH = HCN + O</td>
<td>1.20E+14</td>
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<tr>
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<td>NO + CH$_2$ = HCN + OH</td>
<td>2.00E+13</td>
<td>0.00</td>
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<tr>
<td></td>
<td>NO + CH$_2$ = HCNO + H</td>
<td>2.59E+12</td>
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<td>25.0</td>
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<td></td>
<td>NO + CH$_2$ = HCN + OH</td>
<td>5.01E+11</td>
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<tr>
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<td>NO + CH$_3$ = HCN + H$_2$O</td>
<td>1.50E+12</td>
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<tr>
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<td>NO + CH$_3$ = H$_2$CN + OH</td>
<td>1.00E+12</td>
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<tr>
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<td>NO + CHO = CO + HNO</td>
<td>7.20E+12</td>
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<tr>
<td></td>
<td>NO + C$_3$H = HCN + CO</td>
<td>2.11E+13</td>
<td>0.00</td>
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</tr>
<tr>
<td></td>
<td>NO + HCCO = HCNO + CO</td>
<td>1.30E+13</td>
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### Consumption of N$_2$O

<table>
<thead>
<tr>
<th></th>
<th>N$_2$O + H = OH + N$_2$</th>
<th>9.64E+13</th>
<th>0.00</th>
<th>63.10</th>
</tr>
</thead>
<tbody>
<tr>
<td>37</td>
<td>N$_2$O + O = NO + NO</td>
<td>6.60E+13</td>
<td>0.00</td>
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<tr>
<td></td>
<td>N$_2$O + O = N$_2$O + O$_2$</td>
<td>1.02E+14</td>
<td>0.00</td>
<td>117.0</td>
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<tr>
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<td>N$_2$O + OH = HO$_2$ + N$_2$</td>
<td>2.00E+12</td>
<td>0.00</td>
<td>41.80</td>
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<td>N$_2$O + CO = N$_2$ + CO$_2$</td>
<td>1.25E+12</td>
<td>0.00</td>
<td>72.30</td>
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<td>N$_2$O + CH$_3$ = CH$_3$O + N$_2$</td>
<td>1.00E+15</td>
<td>0.00</td>
<td>119.0</td>
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<td>N$_2$O + M$^<em>$ = O + N$_2$ + M$^</em>$</td>
<td>7.23E+17</td>
<td>-0.73</td>
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### Consumption of NO$_2$

<table>
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<tr>
<th></th>
<th>NO$_2$ + O = NO + O$_2$</th>
<th>1.00E+13</th>
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<th>2.51</th>
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</thead>
<tbody>
<tr>
<td>38</td>
<td>NO$_2$ + H = NO + OH</td>
<td>1.00E+14</td>
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<td>6.27</td>
</tr>
<tr>
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<td>NO$_2$ + N = N$_2$ + O$_2$</td>
<td>1.18E+12</td>
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<td>NO$_2$ + CO = NO + CO$_2$</td>
<td>1.20E+14</td>
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<td>132.0</td>
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<tr>
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<td>NO$_2$ + CH = CHO + NO</td>
<td>5.90E+13</td>
<td>0.00</td>
<td>0.00</td>
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<tr>
<td></td>
<td>NO$_2$ + CH$_2$ = CH$_3$O + NO</td>
<td>5.90E+13</td>
<td>0.00</td>
<td>0.00</td>
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</table>

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<table>
<thead>
<tr>
<th>No.</th>
<th>Reactions</th>
<th>$B_0$</th>
<th>$a_0$</th>
<th>$E_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>NO$_2$ + CH$_3$ = CH$_3$O + NO</td>
<td>1.30E+13</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>2.</td>
<td>NO$_2$ + CHO = CO$_2$ + H + NO</td>
<td>8.40E+15</td>
<td>-0.75</td>
<td>8.07</td>
</tr>
<tr>
<td>3.</td>
<td>NO$_2$ + CHO = CO + HNO</td>
<td>2.10E+00</td>
<td>3.30</td>
<td>9.82</td>
</tr>
<tr>
<td>4.</td>
<td>NO$_2$ + HCCO = NCO + CO + OH</td>
<td>5.00E+12</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>5.</td>
<td>NO$_2$ + HCCO = HNCO + CO$_2$</td>
<td>5.00E+12</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>6.</td>
<td>NO$_2$ + HCCO = HCN + CO$_2$ + O</td>
<td>5.00E+12</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>7.</td>
<td>NO$_2$ + M$^<em>$ = NO + O + M$^</em>$</td>
<td>1.10E+16</td>
<td>0.00</td>
<td>276.0</td>
</tr>
<tr>
<td>8.</td>
<td>NO$_2$ + NO$_2$ = NO + NO + O$_2$</td>
<td>1.60E+12</td>
<td>0.00</td>
<td>109.0</td>
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</table>

**Consumption of HNO**

<table>
<thead>
<tr>
<th>No.</th>
<th>Reactions</th>
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</thead>
<tbody>
<tr>
<td>39.</td>
<td>HNO + H = NO + H$_2$</td>
<td>1.81E+13</td>
<td>1.90</td>
<td>4.16</td>
</tr>
<tr>
<td></td>
<td>HNO + OH = NO + H$_2$O</td>
<td>1.32E+07</td>
<td>1.90</td>
<td>-4.00</td>
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<td></td>
<td>HNO + N = NO + NH</td>
<td>1.00E+13</td>
<td>0.00</td>
<td>8.30</td>
</tr>
<tr>
<td></td>
<td>HNO + O$_2$ = NO + HO$_2$</td>
<td>3.16E+12</td>
<td>0.00</td>
<td>12.5</td>
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<tr>
<td></td>
<td>HNO + NH$_2$ = NO + NH$_3$</td>
<td>5.00E+13</td>
<td>0.00</td>
<td>4.20</td>
</tr>
<tr>
<td></td>
<td>HNO + HNO = N$_2$O + OH</td>
<td>3.90E+12</td>
<td>0.00</td>
<td>209.0</td>
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<tr>
<td></td>
<td>HNO + NO = N$_2$O + H$_2$O</td>
<td>2.00E+12</td>
<td>0.00</td>
<td>109.0</td>
</tr>
<tr>
<td></td>
<td>HNO + NO$_2$ = HNO$_2$ + NO</td>
<td>6.02E+11</td>
<td>0.00</td>
<td>8.31</td>
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<tr>
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<td>HNO + M$^<em>$ = NO + H + M$^</em>$</td>
<td>1.50E+16</td>
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**Consumption of HNO$_2$**

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<thead>
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<th>No.</th>
<th>Reactions</th>
<th>$B_0$</th>
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<th>$E_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>40.</td>
<td>HNO$_2$ + H = NO$_2$ + H$_2$</td>
<td>1.20E+13</td>
<td>0.00</td>
<td>30.7</td>
</tr>
<tr>
<td></td>
<td>HNO$_2$ + O = NO$_2$ + OH</td>
<td>1.20E+13</td>
<td>0.00</td>
<td>25.1</td>
</tr>
<tr>
<td></td>
<td>HNO$_2$ + OH = NO$_2$ + H$_2$O</td>
<td>1.30E+10</td>
<td>1.00</td>
<td>0.56</td>
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</tbody>
</table>

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50 – 55: Reactions of C-H-N-O Species

**Consumption of HCN**

<table>
<thead>
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<th>No.</th>
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<th>$a_0$</th>
<th>$E_0$</th>
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<tbody>
<tr>
<td>50.</td>
<td>HCN + O = NCO + H</td>
<td>1.11E+06</td>
<td>2.10</td>
<td>25.6</td>
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<tr>
<td></td>
<td>HCN + O = NH + CO</td>
<td>2.77E+05</td>
<td>2.10</td>
<td>25.6</td>
</tr>
<tr>
<td></td>
<td>HCN + OH = HNCO + H</td>
<td>4.77E+11</td>
<td>0.00</td>
<td>91.4</td>
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<tr>
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<td>HCN + CN = C$_2$N$_2$ + H</td>
<td>2.00E+13</td>
<td>0.00</td>
<td>0.00</td>
</tr>
</tbody>
</table>

Continued to the next page
## Appendix B

### Consumption of CN/C$_2$N$_2$

<table>
<thead>
<tr>
<th>No.</th>
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<th>$E_\beta$</th>
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</thead>
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<tr>
<td>51</td>
<td>CN + O = CO + N</td>
<td>1.00E+13</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td>CN + OH = NCO + H</td>
<td>6.00E+13</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td>CN + O$_2$ = NCO + O</td>
<td>6.60E+12</td>
<td>0.00</td>
<td>-1.70</td>
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<tr>
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<td>CN + H$_2$ = HCN + H</td>
<td>3.10E+05</td>
<td>2.40</td>
<td>9.30</td>
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<tr>
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<td>CN + H$_2$O = HCN + OH</td>
<td>7.83E+12</td>
<td>0.00</td>
<td>31.1</td>
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<tr>
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<td>CN + N = N$_2$ + C</td>
<td>1.04E+15</td>
<td>-0.50</td>
<td>0.00</td>
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<tr>
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<td>CN + NO = N$_2$ + CO</td>
<td>1.07E+14</td>
<td>0.00</td>
<td>33.4</td>
</tr>
<tr>
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<td>CN + NO = NCO + N</td>
<td>9.64E+13</td>
<td>0.00</td>
<td>176.0</td>
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<td>CN + N$_2$O = NCO + N$_2$</td>
<td>1.00E+13</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td>CN + NO$_2$ = NCO + NO</td>
<td>3.00E+13</td>
<td>0.00</td>
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</tr>
<tr>
<td></td>
<td>CN + CH$_4$ = HCN + CH$_3$</td>
<td>9.03E+12</td>
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<tr>
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<td>C$_2$N$_2$ + O = NCO + CN</td>
<td>4.57E+12</td>
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### Consumption of HNCO/HCNO

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<th>$E_\beta$</th>
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<tr>
<td>52</td>
<td>HCNO + H = HCN + OH</td>
<td>1.00E+14</td>
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<tr>
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<td>HCNO + H = HNCO + H</td>
<td>1.00E+11</td>
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</tr>
<tr>
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<td>HNCO + H = NH$_2$ + CO</td>
<td>2.25E+07</td>
<td>1.70</td>
<td>15.9</td>
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<tr>
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<td>HNCO + O = NH + CO$_2$</td>
<td>9.60E+07</td>
<td>1.40</td>
<td>35.6</td>
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<td>HNCO + O = NCO + OH</td>
<td>2.20E+06</td>
<td>2.10</td>
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<tr>
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<td>HNCO + O = HNO + CO</td>
<td>1.50E+08</td>
<td>1.60</td>
<td>184.0</td>
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<tr>
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<td>HNCO + OH = NCO + H$_2$O</td>
<td>6.40E+05</td>
<td>2.00</td>
<td>10.7</td>
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<td>HNCO + O$_2$ = HNO + CO$_2$</td>
<td>1.00E+12</td>
<td>0.00</td>
<td>146.0</td>
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<td>HNCO + HO$_2$ = NCO + H$_2$O$_2$</td>
<td>3.00E+11</td>
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<td>121.0</td>
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<td>HNCO + M$^<em>$ = NH + CO + M$^</em>$</td>
<td>1.10E+16</td>
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<td>HNCO + NH = NCO + NH$_2$</td>
<td>3.03E+13</td>
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<td>HNCO + NH$_2$ = NCO + NH$_3$</td>
<td>5.00E+12</td>
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### Consumption of NCO

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<th>$E_\beta$</th>
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<td>53</td>
<td>NCO + O = NO + CO</td>
<td>4.20E+13</td>
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<td>0.00</td>
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<tr>
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<td>NCO + H = NH + CO</td>
<td>5.20E+13</td>
<td>0.00</td>
<td>0.00</td>
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</table>

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<table>
<thead>
<tr>
<th>No.</th>
<th>Reactions</th>
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<th>$a_\beta$</th>
<th>$E_\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>244</td>
<td>NCO + OH = CHO + NO</td>
<td>5.00E+12</td>
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<td>62.7</td>
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<tr>
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<td>NCO + H₂ = HNCO + H</td>
<td>7.60E+02</td>
<td>3.00</td>
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<td>NCO + N = N₂ + CO</td>
<td>2.00E+13</td>
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</tr>
<tr>
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<td>NCO + O₂ = NO + CO₂</td>
<td>2.00E+12</td>
<td>0.00</td>
<td>83.6</td>
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<tr>
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<td>NCO + $M^<em>$ = N + CO + $M^</em>$</td>
<td>1.00E+15</td>
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<td>NCO + NO = N₂O + CO</td>
<td>6.20E+17</td>
<td>-1.7</td>
<td>3.19</td>
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<td>NCO + NO = N₂ + CO₂</td>
<td>7.80E+17</td>
<td>-1.7</td>
<td>3.19</td>
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<td>NCO + NCO = N₂ + CO + CO</td>
<td>1.80E+13</td>
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<td>0.00</td>
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<td>NCO + NO₂ = CO + NO + NO</td>
<td>1.30E+13</td>
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</tr>
<tr>
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<td>NCO + NO₂ = CO₂ + N₂O</td>
<td>5.40E+12</td>
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</tr>
<tr>
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<td>NCO + HNO = HCNO + CO</td>
<td>1.80E+13</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td>NCO + HNO₂ = HCNO + CO</td>
<td>3.60E+12</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td>NCO + CHO = HCNO + CO</td>
<td>3.60E+13</td>
<td>0.00</td>
<td>0.00</td>
</tr>
</tbody>
</table>

Consumption of C

| 54  | CH + H = C+ H₂      | 1.50E+14 | 0.00 | 0.00 |
|     | C + O₂ = CO+ O      | 5.00E+13 | 0.00 | 0.00 |
|     | C + NO = CN + O     | 6.60E+13 | 0.00 | 0.00 |

Consumption of H₂CN

| 55  | H₂CN + N = N₂ + CH₂ | 2.00E+13 | 0.00 | 0.00 |
|     | H₂CN + $M^*$ = HCN+ H + $M^*$ | 3.00E+14 | 0.00 | 92.0 |

Arrhenius rate coefficient of the forward reaction, $k_{\beta \beta} = B_\beta T^{a_\beta} \exp (-E_\beta / RT)$ and the molecular density of the third body is $[M^*] = [H₂] + 6.5[H₂O] + 0.4[O₂] + 0.4[N₂] + 0.75[CO] + 1.5[CO₂] + 6.5[CH₄]$. 
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Gills, B. G. (1973). Production and emission of solids, SO\textsubscript{X} and NO\textsubscript{X} from liquid fuel flames. Journal of the Institute of Fuel, 46, 71-76.


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