Diss. ETH Nr. 14492

NO$_x$ Reduction in Partially Premixed Combustion

A dissertation submitted to the
EIDGENÖSSISCHE TECHNISCHE HOCHSCHULE ZÜRICH
for the degree of
Doctor of Technical Sciences

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Zurich, 2001
To my revered Parents
&
my beloved wife, Bhavana
Acknowledgements

Words fell short as I extend my acknowledgements to all those people who make me feel fortunate for where I stand today. To begin with I express my sincere gratitude to Prof. Dimos Poulikakos for his advice and inspirational meetings during my research at ETH, Zurich.

I thank Dr. Juerg Gass for his generous administrative support and for adding fresh perspectives and expertise on the subject of turbulent flames.

I am sincerely thankful to Prof. Ishwar Puri for his inspirational support and series of interactive dialogues. His commitment to the research in Combustion field has always inspired me.

I am thankful to Dr. Tamas Turanyi & Istvan Zsigy from Eoetvoes University, Budapest, for the fruitful collaboration.

I thank all my co-researchers and colleagues at LTNT, especially Andrea Prospe-ro, Andreas Obieglo, Christian Del Taglia, Lale Demiraydin and Lars Blum for all technical and non technical discussion sessions. I am also grateful to Yiannis Ventikos & Rustem Simitovic for their support and valuable feedback.

I thank all the folks at Air and Climate Group, especially Alfred Moser, Daniel Gubler, Peter Roseman, Philip Lengweiler, Stephan Barp, Yuichi Takemasa for the fun filled company.

I thank my friends and CFD colleagues AN Benim (FH Duesseldorf), Ennio Pasquolotto (Wartsila NSD), Geoffrey Engelbrecht, Jorge Ferreira (AEA), Khawar Syed (Alstom) who have played a great role in reminding me of my priorities and for their prompt response to my technical needs.

I extend my heartiest thanks to Late Thomas Gerhardt, who helped me a lot during my initial stay at Stuttgart, Germany and kindled a spirit of optimism in me as my career initiated a new era in foreign land far away from my home.

I thank my parents, sister Snigdha and brother Gaurav for motivating me towards higher achievements in life. Also I could never equate for the patience and sacrifice that my wife, Bhavana has showed. Her love and absolute confidence in me has always bolstered strength and confidence in my work. To them I dedicate my thesis.

I thank Dr. A. Hintermann (Swiss Department of Energy) and The Swiss Federal Institute of Technology for the financial support for the research work.

And last but not least I thank all my desi friends, Anurag, Amit, Samarjit, Shilpee and Suresh for making me feel at home in Zurich.

-Pankaj Bajaj
Zurich, Switzerland
January 2002
Abstract

In the present study the partially premixed combustion is investigated, with its potential to reduce the NO\textsubscript{x} emissions.

The laminar partially premixed flames show a distinct double flame structure under certain boundary conditions of equivalence ratios and strain rates. The double zone structure comprises two separate reaction zones, one rich premixed and the other nonpremixed. If the fuel/air mixture at the rich boundary lies outside the flammability limit, then the mixture is only ignitable if sufficient energy and radical pool is available from the nonpremixed reaction zone. The position of the rich premixed reaction zone depends upon the energy flow from the corresponding nonpremixed zone. Reactants are consumed differently in the two zones. In the rich premixed zone, the methane/air mixture reacts to produce the partially oxidized stable species CO and H\textsubscript{2}. These intermediates are convected towards the nonpremixed zone and oxidized by the oxygen supplied from the lean side. It has been shown that the double zone structure helps in reducing the NO\textsubscript{x} emission as compared to a comparable nonpremixed flame. In the rich premixed reaction region little oxygen is available for NO production through the prompt mechanism. If the rich mixture equivalence ratio is greater than 1.6, then no NO\textsubscript{x} is produced in that region via prompt mechanism. This leads to lower total NO\textsubscript{x} emissions from laminar flames that have a double reaction zone structure as compared with nonpremixed flames. The stabilization of the double flame structure is very sensitive with respect to the equivalence ratio and the strain rate for a counterflow-coflow laminar flames.

The performance of several reaction mechanisms (GRI 2.11, GRI 3.0, CEC and CECR) for methane/air combustion are investigated for the prediction of the above mentioned double flame structure for laminar partially premixed flames. CECR reaction mechanism is able to predict better position of the rich flame in the double flame structure as compared with the predictions of the other mentioned reaction mechanisms. The sensitivity analysis is carried out for the GRI 3.0 reaction mechanism and the suspicious reactions are found that are causing the wrong prediction of the position of the rich premixed flame attached to the diffusion flame. The reaction rates are compared with the NIST experimental data.

These rates are modified for the better prediction of the position of the rich premixed flame in the double flame structure. The modified rates are found to be lying within the experimental uncertainty. In order to justify our modification representative experiments are required for determining the C\textsubscript{2} concentrations in the partially premixed laminar flames. The performance of available reaction mechanisms are investigated for predicting the NO\textsubscript{x} emissions in the laminar partially premixed flames. None of the mechanism is able to predict the NO\textsubscript{x} concentration correctly. The modified GRI 3.0 reaction mechanism gives better prediction of the NO concentrations as compared with the predictions of the other mechanisms in the partially premixed laminar counterflow flames. The reasons for incorrect prediction of the NO emissions lie in the incorrect prediction of the CH concentration, which plays an important role in the initiation of prompt mechanism for NO\textsubscript{x} pro-
duction. However, there are very few experiments carried out in determining the CH concentration with laser diagnostics methods in the laminar rich partially premixed flames showing double flame structure. So new experiments are needed for laminar partially premixed flames to measure the CH concentration in the rich premixed region of the double flame structure. This would help in further development of these mechanisms to predict NO\textsubscript{x} correctly.

In the present work, the in-house built transient laminar flamelet model (TLFM) is also tested for the prediction of the partially premixed turbulent flame. This model is used to predict the extinction/re-ignition phenomenon in the turbulent diffusion flames. The potential and the weakness of the existing model are evaluated.

The turbulent diffusion flames can be described with the two variables, namely the mixture fraction and the scalar dissipation rate, in the flamelet approach of the combustion modeling. On the other hand the structure of the premixed turbulent flames are predicted well using variables, the reaction progress variable and the strain rate (scalar dissipation rate) in the flamelet models. So in order to describe the partially premixed turbulent flames using the fast chemistry approach, the important features of flames can be captured by determining the relationship of all the scalars as a function of the mixture fraction, the reaction progress variable and the scalar dissipation rate.

The in-house built TLFM uses all these three variables, namely the mixture fraction, the reaction progress variable and the scalar dissipation rate, for describing the turbulent diffusion flames with very high Reynolds number.

The range of the reaction progress variable in order to predict the re-ignition phenomenon in the diffusion flame cases is assumed in the flamelet libraries. This range is from 0.5 till 1.0 and is able to capture the re-ignition processes.

The existing TLFM with transient laminar flamelet libraries is unable to capture the flame structure for turbulent partially premixed flames due to this insufficient range for the reaction progress variable. The new transient laminar flamelet libraries are developed covering the whole range of reaction progress variable from 0 till 1 with steps of 0.1. Moreover, the flamelet model is modified by using the transformation of the mixture fraction space into the normalised mixture fraction space (value between 0 and 1) of the diffusion flame. In this way the partially premixed flame in the turbulent flame compares reasonably well with the diffusion flame structure in the mixture fraction space of the flamelet libraries. The important features of the turbulent partially premixed flame, i.e. maximum temperature, flame length, species concentrations, is captured with the existing TLFM using new transient flamelet libraries.

The effect of the partial premixing is also observed in the numerical calculation of the test flame (partially premixed in nature) with different equivalence ratios in the main jet stream. The flame length decreases with the increase in the partial premixing. These observations are also found in the experiments on the ELCO household burner, which was operated, in the partially premixed mode.

This model shows promising results and can be used for the design of practical burners, operated in partially premixed modes. However, further improvement of
the predictive potential are needed with respect to the prediction of the interme-
diate product species (H₂ and CO).
Zusammenfassung

In dieser Studie wird die teilvorgemischte Verbrennung und deren Potential zur Reduktion der NOx Emissionen erforscht. Bei bestimmten Äquivalenzverhältnissen und Streckungsrate zeigen die laminären teilvorgemischten Flammen eine deutliche doppelte Flammenstruktur. Diese doppelte Flammenstruktur besteht aus zwei getrennten Reaktionszonen, einer vollvorgemischten einerseits und einer nichtvorgemischten andererseits. Wenn die Brennstoff/Luft-Mischung an der fetten vollvorgemischten Grenze außerhalb der Zündgrenze liegt, ist die Mischung nur entzündbar, falls genügend Energie und genügend Radikale von der nichtvorgemischten Reaktionszone vorhanden sind. Die Position der vollvorgemischten Zone hängt von der Energie ab, die von der entsprechenden nichtvorgemischten Zone abfließt. Die Reaktanden werden in den zwei Zonen verschieden konsumiert. In der vollvorgemischten Zone reagiert die Methan/Luft-Mischung zu den teilweise oxidierten stabilen Spezies CO und H2. Diese Zwischenprodukte werden durch Konvektion in die nichtvorgemischte Zone transportiert und oxidieren dort mit Hilfe des Sauerstoffs, der von der mageren Seite geliefert wird. Es wurde gezeigt, dass die doppelte Flammenstruktur mithilft, die doppelte Flammenstruktur in Teilvorgemischten laminären Flammen zu reduzieren. In der fetten vollvorgemischten Region ist nur wenig Sauerstoff für NOx Produktion aufgrund des prompten Mechanismus vorhanden. Wenn in der fetten Mischung das Äquivalenzverhältnis größer als 1,6 ist, wird kein NOx in dieser Region über den Prompt Mechanismus produziert. Dies führt zu tieferen totalen NOx Emissionen von laminären Flammen mit doppelter Reaktionszone im Vergleich zu nichtvorgemischten Flammen. Die Stabilisierung der doppelten Flammen-Struktur ist in bezug auf das Äquivalenzverhältnis und die Streckungsrate für die counterflow-/coflow laminären Flamme sehr sensitiv.

Die Leistung der verschiedenen Reaktionsmechanismen (GRI 2.11, GRI 3.0, CEC und CECR) für die Methan/Luft Verbrennung wurde analysiert, um bessere Vorhersagen der doppelten Flammenstruktur in teilvorgemischten laminären Flammen zu gewährleisten. Der CECR Reaktionsmechanismus ist besser fähig, die Position der fetten Flamme in der doppelten Flammenstruktur vorhersagen als die anderen erwähnten Reaktionsmechanismen. Eine Sensitivitätsanalyse wurde für den GRI 3.0 Reaktionsmechanismus durchgeführt und die fraglichen Reaktionen gefunden, welche die falsche Vorhersage der Position der fetten vollvorgemischten Flamme in Verbindung mit der Diffusionsflamme verursachen. Die Reaktionsraten wurden mit den experimentellen Daten in der NIST Datenbank verglichen.

Diese Raten werden zur besseren Vorhersage der Position der fetten vollvorgemischten Flamme in der doppelten Flammenstruktur modifiziert. Die Werte für die modifizierten Raten liegen innerhalb der experimentellen Ungewissheit. Um diese Modifikation zu rechtfertigen werden repräsentative Experimente für die Bestimmung der C2 Konzentrationen in teilvorgemischten laminären Flammen benötigt. Für die Vorhersage der NOx Emissionen in laminären teilvorgemischten
Flammen wurde die Leistungsfähigkeit der vorhandenen Reaktionsmechanismen untersucht. Keiner der Mechanismen ist fähig, die NOₓ Konzentration korrekt vorherzusagen. Der modifizierte GRI 3.0 Reaktionsmechanismus produziert bessere Vorhersagen von NOₓ Konzentrationen im Vergleich mit anderen Mechanismen für die teilvorgemischte laminare counterflow Flamme. Die Gründe für die falsche Vorhersage der NOₓ Emissionen liegen in der falschen Vorhersage der CH Konzentration, die eine wichtige Rolle in der Einleitung des prompten Mechanismus für NOₓ Produktion spielt. Es gibt nur sehr wenige Experimente, die mithilfe von Laserdiagnosemethoden die CH Konzentration in laminar teilvorgemischten Flammen mit doppelter Flammenstruktur bestimmt haben. Solche neue Experimente werden gebraucht für die Messung der CH Konzentration in laminar teilvorgemischten Flammen mit doppelter Flammenstruktur. Diese wären hilfreich für die Weiterentwicklung der Mechanismen zur Vorhersage der NOₓ Konzentration.

In dieser Arbeit wird ebenfalls das selbsterstellte "Transient Laminar Flamelet Modell" (TLFM) für die Vorhersage der teilvorgemischten turbulenten Flamme getestet. Dieses Modell wird normalerweise benutzt, um Verlösungs-/Wiederzündungs-Phänomene in der turbulenten Diffusionsflamme vorherzusagen. Die Möglichkeiten und Schwächen des bestehenden Modells für die Vorhersage der turbulenten teilvorgemischten Flamme werden abgeschätzt.

Die turbulente Diffusionsflamme kann in diesem Modell mit Hilfe von zwei Variablen, nämlich dem Mischungsbruch und der skalaren Dissipation, beschrieben werden. Andererseits wird die Struktur der vorgemischten turbulenten Flamme gut unter Benutzung der Variablen Reaktionsfortschritt und skalare Dissipation vorgehersagt. Es genügt also, um die turbulente teilvorgemischte Flamme mit Hilfe der Annahme schneller Chemie zu beschreiben, dass die wichtigen Merkmale der Flamme durch die Beschreibung aller Skalare als Funktionen des Mischungsbruchs, des Reaktionsfortschritt und des Streckungsverhältnisses erfasst werden.

Das TLFM benutzt alle drei Variablen, also den Mischungsbruch, den Reaktionsfortschritt und das Streckungsverhältnis, um die turbulente Diffusionsflamme für sehr hohe Reynoldszenahlen zu beschreiben.

Der Wertebereich der Variablen für den Reaktionsfortschritt für die Vorhersage des Wiederentzündungsphänomens in der Diffusionsflamme wird mithilfe einer Bibliothek bestimmt. Dieser Wertebereich reicht von 0.5 bis 1.0 und ist gut geeignet, Wiederentzündungsprozesse abzubilden.

Das bestehende TLFM mit der transienten Flamelet Bibliothek ist aufgrund des falschen Wertebereiches nicht geeignet, die Flammenstruktur für die teilvorgemischte turbulente Flamme zu beschreiben. Eine neue Flamelet Bibliothek wurde entwickelt, die den ganzen Wertebereich für den Reaktionsfortschritt Variable von 0 bis 1 in Teilschritten von 0.1 abdeckt. Darüber hinaus wurde das Flamelet Modell durch die Verwendung der Transformation des Mischungsbruches in den normalisierten Mischungsbruchraum (Wertebereich 0 bis 1) abgeändert. Auf diese Weise stimmt die teilvorgemischte turbulente Flamme gut mit der Diffusionsflammenstruktur im Wertebereich der Flamelet Bibliothek überein. Die wichtigen Merkmale der turbulenten teilvorgemischten Flamme, zum Beispiel die maximale
Temperatur, die Flammenlänge und die Spezies-Konzentrationen, werden mit dem bestehenden TLFM und der neuen transienten Bibliothek gut erfasst.


Dieses Modell zeigt vielversprechende Ergebnisse, und kann benutzt werden für den Entwurf von praktischen Brennern, die im teilvorgemischten Zustand betrieben werden. Weitere Verbesserungen werden aber in bezug auf bessere Vorhersagen der zwischenprodukte (H₂ und CO) benötigt.
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<th>Units</th>
<th>Description</th>
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</thead>
<tbody>
<tr>
<td>A, B, C</td>
<td>-</td>
<td>model constants</td>
</tr>
<tr>
<td>A</td>
<td>mole, cm$^3$s</td>
<td>pre-exponential factor of rate constant</td>
</tr>
<tr>
<td>B</td>
<td>-</td>
<td>overall sensitivity</td>
</tr>
<tr>
<td>C</td>
<td>-</td>
<td>turbulence model constants</td>
</tr>
<tr>
<td>$C_{EBU}$</td>
<td>-</td>
<td>model constants</td>
</tr>
<tr>
<td>D</td>
<td>m$^2$/s</td>
<td>diffusion coefficient</td>
</tr>
<tr>
<td>Da</td>
<td>-</td>
<td>Damköhler number</td>
</tr>
<tr>
<td>E</td>
<td>kJ/kmol</td>
<td>activation energy</td>
</tr>
<tr>
<td>F, f</td>
<td>-</td>
<td>fuel</td>
</tr>
<tr>
<td>H</td>
<td>J</td>
<td>enthalpy</td>
</tr>
<tr>
<td>J</td>
<td>kg/(m$^2$s)</td>
<td>diffusion flux term</td>
</tr>
<tr>
<td>K</td>
<td>1/s</td>
<td>global strain rate</td>
</tr>
<tr>
<td>L, l</td>
<td>m</td>
<td>characteristic length</td>
</tr>
<tr>
<td>L</td>
<td>m</td>
<td>visible flame length</td>
</tr>
<tr>
<td>N</td>
<td>-</td>
<td>number of moles</td>
</tr>
<tr>
<td>N</td>
<td>-</td>
<td>scalar dissipation rate</td>
</tr>
<tr>
<td>O</td>
<td>-</td>
<td>oxidizer</td>
</tr>
<tr>
<td>P</td>
<td>-</td>
<td>product</td>
</tr>
<tr>
<td>$P_{pc}$</td>
<td>m$^2$/s$^3$</td>
<td>pope correction term</td>
</tr>
<tr>
<td>P</td>
<td>*</td>
<td>probability density function</td>
</tr>
<tr>
<td>S</td>
<td>-</td>
<td>normalized sensitivity</td>
</tr>
<tr>
<td>$S'$</td>
<td>*</td>
<td>chemical source term</td>
</tr>
<tr>
<td>$S_l$</td>
<td>m/s</td>
<td>flame front velocity</td>
</tr>
<tr>
<td>T</td>
<td>K</td>
<td>temperature</td>
</tr>
<tr>
<td>$V_k$</td>
<td>m/s</td>
<td>diffusion velocity</td>
</tr>
<tr>
<td>W</td>
<td>kg/mol</td>
<td>molecular weight</td>
</tr>
<tr>
<td>X</td>
<td>-</td>
<td>mole fraction</td>
</tr>
<tr>
<td>Y</td>
<td>-</td>
<td>mass fraction</td>
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<tr>
<td>Z</td>
<td>-</td>
<td>mixture fraction</td>
</tr>
<tr>
<td>a</td>
<td>1/s</td>
<td>air side strain rate</td>
</tr>
</tbody>
</table>
c – reaction progress variable
J/(kgK) – heat capacity
mm – diameter
m²/s² – turbulent kinetic energy
1/s – reaction rate
kg – mass
r, s – stoichiometric coefficient
s – time
m/s – velocity component
m – space coordinate

* depending on the chosen scalar

### Greek Symbols

<table>
<thead>
<tr>
<th>Symbols</th>
<th>Units</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>α</td>
<td>W/(m²K)</td>
<td>=λ/(ρcₚ), thermal diffusivity</td>
</tr>
<tr>
<td>α, β, Γ</td>
<td>–</td>
<td>empirical constant</td>
</tr>
<tr>
<td>β</td>
<td>–</td>
<td>conserved scalar</td>
</tr>
<tr>
<td>β</td>
<td>–</td>
<td>temperature exponent of the rate constant</td>
</tr>
<tr>
<td>ε</td>
<td>m²/s³</td>
<td>dissipation rate</td>
</tr>
<tr>
<td>η</td>
<td>–</td>
<td>conditional mixture fraction</td>
</tr>
<tr>
<td>λ</td>
<td>–</td>
<td>local air excess ratio</td>
</tr>
<tr>
<td>λ</td>
<td>W/(Km)</td>
<td>heat conductivity</td>
</tr>
<tr>
<td>μ</td>
<td>kg/(ms)</td>
<td>=ρν, dynamic viscosity</td>
</tr>
<tr>
<td>υ₁</td>
<td>kg/(ms)</td>
<td>turbulent viscosity</td>
</tr>
<tr>
<td>ν</td>
<td>m²/s</td>
<td>=μ/ρ, kinematic viscosity</td>
</tr>
<tr>
<td>ν'</td>
<td>–</td>
<td>stoichiometric coefficients forward reaction</td>
</tr>
<tr>
<td>ν''</td>
<td>–</td>
<td>stoichiometric coefficients backward reaction</td>
</tr>
<tr>
<td>ρ</td>
<td>kg/m³</td>
<td>density</td>
</tr>
<tr>
<td>σ</td>
<td>–</td>
<td>model constant</td>
</tr>
<tr>
<td>ξ</td>
<td>–</td>
<td>modified mixture fraction</td>
</tr>
<tr>
<td>τ</td>
<td>s</td>
<td>characteristic time</td>
</tr>
<tr>
<td>φ</td>
<td>–</td>
<td>local equivalence ratio</td>
</tr>
<tr>
<td>φ</td>
<td>–</td>
<td>global mass flow equivalence ratio</td>
</tr>
</tbody>
</table>
\( \Phi \) - scalar variable
\( \Phi \) - thermochemical vector
\( \Psi \) - vector of independent variables in stated space
\( \chi \) - 1/s scalar dissipation rate
\( \zeta \) - premixedness parameter
\( \omega \) - source term

### Subscripts

<table>
<thead>
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<td>adia</td>
<td>adiabatic conditions</td>
</tr>
<tr>
<td>F,f</td>
<td>fuel</td>
</tr>
<tr>
<td>fl</td>
<td>flamelet</td>
</tr>
<tr>
<td>i</td>
<td>species number</td>
</tr>
<tr>
<td>j</td>
<td>reaction number</td>
</tr>
<tr>
<td>L</td>
<td>lean mixture</td>
</tr>
<tr>
<td>m</td>
<td>number of carbon atoms in hydrocarbon species</td>
</tr>
<tr>
<td>min</td>
<td>minimum</td>
</tr>
<tr>
<td>max</td>
<td>maximum</td>
</tr>
<tr>
<td>n</td>
<td>number of hydrogen atoms in hydrocarbon species</td>
</tr>
<tr>
<td>o</td>
<td>oxidizer</td>
</tr>
<tr>
<td>p</td>
<td>product</td>
</tr>
<tr>
<td>p</td>
<td>pressure</td>
</tr>
<tr>
<td>R</td>
<td>rich mixture</td>
</tr>
<tr>
<td>r</td>
<td>reactants</td>
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<td>st</td>
<td>stoichiometric</td>
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<td>t</td>
<td>turbulent</td>
</tr>
<tr>
<td>u</td>
<td>unburnt stage</td>
</tr>
<tr>
<td>( \varepsilon )</td>
<td>dissipation</td>
</tr>
<tr>
<td>( \mu )</td>
<td>model constant</td>
</tr>
<tr>
<td>( O_2 )</td>
<td>oxidizer</td>
</tr>
<tr>
<td>( CO_2 )</td>
<td>carbondioxide</td>
</tr>
<tr>
<td>( H_2O )</td>
<td>water</td>
</tr>
<tr>
<td>1</td>
<td>fuel inlet stream</td>
</tr>
<tr>
<td>2</td>
<td>oxidizer inlet stream</td>
</tr>
</tbody>
</table>
1 \text{ local values}

**Superscripts**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>-</td>
<td>time averaged, reynolds averaged</td>
</tr>
<tr>
<td>~</td>
<td>favre averaged</td>
</tr>
<tr>
<td>.</td>
<td>production rate</td>
</tr>
<tr>
<td>&quot;2</td>
<td>variance of the scalar</td>
</tr>
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</table>
1 Introduction

The research for purification of the atmospheric air has received much more attention in recent years. Although there has been significant improvement in the quality of air in some of the European cities in the last decade, air pollution is still a very serious concern.

Table 1.1 shows the "Europe" study done by Darmstadter et al. [36] in 1987. They used a variety of computer models and evaluation techniques to derive predicted concentrations of selected atmospheric parameters. In their study, the four important atmospheric properties, namely, photochemical smog, acid precipitation, corrosion of metals, and stratospheric ozone were selected for analysis for the epochs 1890, 1920, 1950, 1980, 2030, and 2080. These properties were selected for their direct impact on the quality of living in the urban cities. Such a study can help in assessing the need for technological development in the combustion industries that are the root cause for emissions of the major air pollutants. It can also help in showing us the directions for the future research in the combustion technology.

Table 1.1: Environmental Quality Assessments for Europe.

<table>
<thead>
<tr>
<th>averaged values over the year</th>
<th>1890</th>
<th>1920</th>
<th>1950</th>
<th>1980</th>
<th>2030C</th>
<th>2080C</th>
<th>2030D</th>
<th>2080D</th>
</tr>
</thead>
<tbody>
<tr>
<td>O3, ppb</td>
<td>10.0</td>
<td>20.0</td>
<td>35.0</td>
<td>55.0</td>
<td>180.0</td>
<td>280.0</td>
<td>130.0</td>
<td>130.0</td>
</tr>
<tr>
<td>Smog severity</td>
<td>L</td>
<td>L</td>
<td>L/M</td>
<td>H</td>
<td>H</td>
<td>H</td>
<td>H</td>
<td>H</td>
</tr>
<tr>
<td>Precipitation pH</td>
<td>5.4</td>
<td>5.4</td>
<td>5.3</td>
<td>4.3</td>
<td>3.5</td>
<td>2.8</td>
<td>3.8</td>
<td>3.3</td>
</tr>
<tr>
<td>Precipitation acidity</td>
<td>L</td>
<td>L</td>
<td>L</td>
<td>M</td>
<td>H</td>
<td>H</td>
<td>H</td>
<td>H</td>
</tr>
<tr>
<td>SO2, ppb</td>
<td>7.0</td>
<td>11.0</td>
<td>12.0</td>
<td>14.0</td>
<td>40.0</td>
<td>160.0</td>
<td>25.0</td>
<td>60.0</td>
</tr>
<tr>
<td>Corrosion severity</td>
<td>L/M</td>
<td>M</td>
<td>M</td>
<td>M</td>
<td>M/H</td>
<td>H</td>
<td>M</td>
<td>M/H</td>
</tr>
</tbody>
</table>

KEY: C-Constant-emissions scenario; D-declining-emissions scenario; L-low impact; M-moderate impact; and H-high impact.

Darmstadter et al. used two development scenarios for the emission predictions. These scenarios are briefly summarized here. The first scenario, namely C in reference to its constant emission coefficients, is a conventional growth scenario for the world. It predicts global growth rates of 1.5, 3.2, and 2.1 percent per year for population, gross national product, and total energy use, respectively, for the period 1980-2030. For the period 2030-2080 the comparable growth rates predicted are 0.3, 2.2, and 1.8 percent per year. Also appropriate growth rates are assigned to particular geographical regions of interest for each period. The emission factors, which are defined as the mass of emittants per unit mass of feed stock in a given source process, are assumed to remain constant at 1980 values.

The second growth scenario, termed as scenario D in reference to its decreasing emission coefficients, is identical to the first in its global and regional growth rates. It differs from scenario C only in that it assumes that the emission factors for the...
Introduction

sources will decrease over the century 1980-2080 at a rate of 1 percent per year. This scenario, therefore, predict that increases in the total use of feed stocks will be offset by improvements in technology.

However, it can be observed from both these scenarios (cf. columns 2030C, 2030D in the Table 1.1) that the impact of the pollutants will be high in the future. The swiss pollution laws [67] enforced from July 1993 gives 78 ppb of SO$_2$ concentration averaged over the whole year and 196 ppb of O$_3$ averaged over a month as the upper limit for the pollutants. There is a need to accelerate the research in order to reduce the conventional pollutant emissions to the lowest achievable levels in order to breathe in a pollution free atmosphere.

The two most important oxides of nitrogen with regard to air pollution are nitric oxide (NO) and nitrogen dioxide (NO$_2$), jointly referred to as NO$_x$ in the context of the environment. Nitrogen dioxide (NO$_2$) is a highly reactive gas that is readily formed through the combination of nitric oxide (NO) with oxygen. This reaction is typically a natural process. The production of the nitrogen oxide by natural processes accounts for about 30% of the total NO$_x$ production. It occurs through lightning, volcanic activity, bacterial action in soil and forest fires. The remainder 70% of this pollutant is formed due to human activities [28]. The combustion of fossil fuels by automobiles, power plants, industry and household activities contribute to their concentrations in the environment. NO$_x$ emissions depend strongly on the combustion temperature and they are emitted in very high temperature combustion processes. More than half the nitrogen oxides are produced by stationary fuel combustion sources such as coal or gas burning power plants and industrial boilers. The transportation industry, mainly motor vehicles, contribute about 40% of the load as shown in the following Figure 1.1.

![Figure 1.1 Different Contribution for the NO$_x$ emissions (source Volvo 1996).](image-url)
It is of concern to have high levels of NO\textsubscript{x} in the environment since they combine with volatile organic compounds (VOCs) in the presence of sunlight to form tropospheric ozone. This process contributes to the formation of urban smog. The word smog means contraction of smoke and fog. It refers to an atmospheric haze produced by pollutants. It can be observed from the above "Europe" study (cf. Table 1.1) that the impact of the smog will be quite high in scenario C. Also the scenario D predicted increase in smog concentrations.

Nitrogen oxides also play a major role in atmospheric photochemical reactions that contribute to acid rain. So these gases affects the environment through acidification, forest damage, smog formation, depletion of the stratospheric ozone layer and the greenhouse effect.

Emission standards for 'conventional' pollutants are becoming increasingly stringent e.g. the Swiss pollution laws [67] enforced from July 1993 gives 30 μg/m\textsuperscript{3} of NO\textsubscript{x} concentration averaged over the whole year or 80μg/m\textsuperscript{3} averaged over 24 hours as the upper limit for the NO\textsubscript{x} emissions. Different research groups from all over the world are involved in finding out ways to have better combustion systems producing lesser amount of emissions.

In our opinion the research direction in order to reduce the emissions from the combustion units can be followed in two ways. One effective way to reduce the NO\textsubscript{x} emissions is to treat the exhaust gases coming out of combustion systems. There has been a tremendous amount of research carried out in this field and there are equipments available [66], which can reduce the NO\textsubscript{x} emissions to a level less than 5ppm e.g. in the gas turbine burners. However these techniques are quite expensive and owing to their higher procurable costs, it is not feasible to use such "exhaust gases treatment equipments" for energy processes.

The other way is to further develop the combustion systems by optimizing the combustion processes and the aerodynamics of the combustion unit in order to achieve the minimum possible emissions without sacrificing the efficiency of the system. The majority of industrial gas turbines are operated in lean premixed mode to achieve the required lower emission standards [87]. Engelbrecht et al. [23] have investigated different gas turbine burners that are operated in lean premixed combustion to achieve lower emissions. There is a trend to use natural gas fuel as compared to other conventional fossil fuels like diesel etc. in the industrial burners with an aim to achieve lower conventional air pollutants. The natural gas reserves are abundant as compared to the other fossil fuels [70], which justifies the research on such fuels to be used in the industrial burners. Blum et al. [16] have performed experiments on a commercial household burner [64] that is operated under partially premixed conditions. They used methane gas as fuel for the burner and found that the NO\textsubscript{x} emissions can be reduced substantially by switching the combustion operation from the diffusion to the partial premixed mode.

The rapid improvement in the field of computer technology has lead to a fast progress in understanding turbulent combustion as it occurs in most of the existing combustion devices. However, a principal problem in turbulent reacting flows is the large range of time and length scales which can not be resolved with today's available computer facilities. So to investigate such type of flows, models for describing the turbulent flow and the combustion process are still needed.
Computational fluid dynamics (CFD) plays an important role for designing new combustion units in shortest possible time. There are a number of commercial CFD tools available that can be used for the simulation of practical combustion devices in order to facilitate their further development. These commercial packages [62, 65] contain a number of generic turbulence and combustion models. Our combustion group at the Swiss Federal Institute of Technology (ETH) is involved in the development of complex and detailed combustion models [24, 44]. These models are coupled with the commercial CFD-software [62].

Although there is continuous research going on for the further development of the available models for the exact prediction of the flow field, the heat release and the pollutant species in complex industrial burners still it is possible to use the available models to understand the physics of the combustion units with fair accuracy. CFD helps in designing of the practical combustion units with a smaller amount of resources as compared with the experimental investigation technique. However, experiments are also required for the validation of the developed models and these models can then be used for the designing new energy generation devices.

Hence, the combination of modern experimental techniques such as laser diagnostics with computational analysis, is a powerful tool for the research of reacting and non-reacting flows.

### 1.1 Aims and Objectives

The new technological design of burners in order to get the lowest possible emissions without sacrificing the efficiency is very challenging. The aim of the research project is to investigate the combustion phenomenon in the simple geometry for their potential to reduce the NO emissions.

Industrial burners are typically categorized into nonpremixed (diffusion) and premixed burning modes. However, partially premixed combustion is known to occur in both highly turbulent diffusion flames as well as in technical realizations of premixed burners. Consequently, in these systems the fuel stream becomes enriched with air (and vice-versa) to levels at which the mixture would be barely or not at all ignitible. Under certain conditions, the subsequent combustion occurs under rich premixed conditions that include the corresponding NOx reduction potential, however, without the safety concerns associated with fuel–air premixing. Blum et al. [16] have measured NOx and CO emissions in a commercial household burner, which is operated in the partially premixed mode. This burner is normally operated in a diffusion mode. A reduction of about 35% for the NOx and of 60% for the CO emissions were observed for a high level of premixing of the rich flow. This experimental work gives the motivation to investigate such partially premixed flames.

In the following work presented, the structure of the laminar partially premixed counterflow flames was studied with methane as the fuel. The natural gas contains about 94% - 96% of methane [63]. We tested the methane/air mixture for understanding the partially premixed combustion phenomenon. We also discussed the different mechanisms for the methane/air combustion that can have
Introduction

application in the designing of practical combustion systems. The performance of these reaction mechanisms for the description of the structure of the partially premixed flames and also their predictions for the NO emissions are discussed in this work.

So the aim of the presented research is to understand the structure of the partially premixed combustion phenomenon and to study the potential to reduce the NO emissions.

1.2 Outline

The presented work is divided into three main sections. The first section discusses the different modes of combustion and their industrial application. The progress of the research in the development of different models to describe these modes of turbulent combustion are also explained. The capabilities and the limitations of these models are discussed with respect to the correct predictions of the flame structure and the emissions in the laboratory flames and industrial burners. Some basic definitions that are required in order to understand the interaction of the combustion and the turbulence are also explained.

In the second section we discuss the structure of the laminar partially premixed flames and their potential to reduce the NO emissions. The effects of the different variables, namely strain rate, equivalence ratio, are discussed with respect to the structure of the partially premixed flames. The reaction mechanisms available for the methane/air combustion are discussed with respect to their performance for the prediction of the flame structure and the NO emissions. The limitations of these mechanisms are discussed and the certain reactions are found in order to rectify the limitations of the reaction mechanisms of methane/air combustion in the prediction of the flame structure for particular configurations. The one dimensional code is also described, which computes the steady state solution for axisymmetric flames established between two opposed partially premixed flows.

The third section is dealing with the turbulent partially premixed flames. The transient flamelet model for diffusion flames that is developed in our laboratory is discussed for its potential to describe the partially premixed turbulent flames. This model is also described in this section. A test case is simulated using the transient state flamelet model and the computed results are compared with the available experimental data. The effect of the equivalence ratio on the structure of the partially premixed turbulent flame is also investigated using the flamelet model coupled with the commercial CFD software [62].

The thesis is summarized in the last chapter with an outlook for the research that could be carried out in the direction of further development of the transient flamelet model for the prediction of the turbulent partially premixed combustion mode in the industrial application. The requirement of doing new experiments for measuring certain species, which play an important role in the description of the flame structure and the prediction of the NO emissions in the laminar partially premixed flames are also discussed in the presented thesis.
2 Types of Combustion and their Applicability

There are two classical modes of combustion namely nonpremixed and premixed combustion. However, the partially premixed combustion is the phenomenon that occurs both in the conventional combustion modes in industrial devices. In specific furnaces, the fuel/air is also partly mixed prior to combustion in order to have partially premixed flames. These different modes of combustion and the widely used computational models for their predictions are discussed later after the description of the nomenclature of the terms that are used in this section.

The most widely used combustion models for the computations of the diffusion, the premixed and the partially premixed flames are described in this section to give an overview about these models. These models are used quite often in designing the practical burners. The models would not be discussed in detail and it is suggested to go through the stated references in the section for detailed explanation of the discussed models.

2.1 Description of relevant concepts

The turbulent reacting flows involve the complex interactions of the microscopic/macroscopic mixing of the fluid elements and the chemical reactions. The description of these interactions can be simplified by the use of non-dimensional scalars, namely the mixture fraction and the reaction progress variable. These scalars are discussed below.

2.1.1 Mixture Fraction

In the process of diffusion, mixing plays an important role prior to combustion in diffusion and partially premixed cases. Moreover, during the combustion process, a number of intermediate stable species are generated and also consumed. Their local concentration depends drastically on the mixing process. So it is significant to describe the mixing in terms of some non-dimensional correlation. This term is known as mixture fraction.

The mixture fraction may be defined as the fraction of the mass which comes from the fuel stream. Mixture fraction is a conserved quantity i.e. it is independent of the reaction in the combustion process. It is a conserved scalar, whose actual value at each point is conserved within a flow field. The mixture fraction may be defined by the concentration of the elements in the mixture. Mathematically it may be written as shown below:

\[ Z_j = \sum_{n=1}^{N} \frac{W_n a_n}{W_j} Y_n \]  \hspace{1cm} (2.1)
where \( a_{nj} \) represents the number of elements \( j \) in the species \( n \) and \( W_j, W_n \) are the corresponding molecular weights.

The mixture fraction can take a value of 0 in the pure air stream and 1 in the pure fuel stream. This general expression can be used to find out the relationship between the fuel and the air with the mixture fraction. In this manner the equation for the stoichiometric mixture fraction can be derived. The fuel-air mixture for hydrocarbon fuels is stoichiometric, if the fuel to oxygen ratio is such that both are entirely consumed when the combustion is complete, giving rise to products of \( \text{CO}_2 \) and \( \text{H}_2\text{O} \). This is one of the condition for the air/fuel mixture to be satisfied for the combustion of the non-premixed flames.

The global reaction describing the combustion of any single component of the hydrocarbon fuel may be expressed as

\[
v' F \cdot C_m H_n + v'_O \cdot O_2 = v'' \cdot \text{CO}_2 + v''' \cdot \text{H}_2\text{O}
\]

where the stoichiometric coefficients are

\[
v'_F = 1, \quad v'_O = m + \frac{n}{4}, \quad v'' = m, \quad v''' = \frac{n}{2}
\]

where \( v'_F \) may be chosen arbitrarily to unity. The ratio of the number of moles of fuel and oxidizer in the unburnt mixture should be equal to the ratio of the stoichiometric coefficients for the complete consumption of the one mole of the hydrocarbon fuel.

In terms of mass fractions the relationship can be expressed as

\[
\frac{Y_{\text{O}_2}}{Y_{\text{F},u}} = \frac{W_{\text{O}_2}v'_O}{W_Fv'_F} = v
\]

where \( v \) is called as the stoichiometric mass fraction and \( W \) denotes the molecular weight.

Since the total mass in the system is independent of the chemical reaction, the mass fractions of unburnt state and any later combustion state can be written as follows

\[
\frac{Y_{\text{O}_2} - Y_{\text{O}_2,u}}{W_{\text{O}_2}v'_O} = \frac{Y_{\text{F},u} - Y_{\text{F},u}}{W_Fv'_F}
\]

This can be expressed using the stoichiometric mass fraction as

\[
v_Y F - Y_{\text{O}_2} = v_Y F,u - Y_{\text{O}_2,u}
\]

Also the mass fraction \( Y_{\text{F},u} \) of the fuel in the mixture is proportional to the mass fraction in the original fuel stream, so using the eq. 2.1 one gets
where the $Y_{F,l}$ represents the mass fraction of the fuel in the fuel stream. In a similar manner, one can relate the oxidizer mass fraction with the mixture fraction as shown below

$$Y_{O_2,u} = Y_{O_2,2}(1-Z)$$

(2.7)

where the $Y_{O_2,2}$ denotes the mass fraction of oxygen in the oxidizer stream.

Introducing the values of unburnt fuel and oxidizer mass fraction from eq. 2.6 and 2.7 into eq. 2.5, one gets

$$Z = \frac{\nu Y_{F,u} - Y_{O_2} + Y_{O_2,2}}{Y_{O_2,2} + \nu Y_{F,l}}$$

(2.8)

The above expression implies that mixture fraction is bounded between 0 and 1 i.e. it attains the value unity in pure fuel stream and zero in pure oxidizer stream. So the mixture fraction does not describe whether the fuel is burnt or not. It is directly related to the mixing phenomenon.

One obtains the relation $\nu Y_{F} = Y_{O_2}$ at the stoichiometry, so the expression for the stoichiometric mixture fraction is as follows

$$Z_{st} = \left[1 + \frac{\nu Y_{F,l}}{Y_{O_2,2}}\right]^{-1}$$

(2.9)

Another term analogous to mixture fraction, which is often used in describing the boundary conditions of combustion processes is equivalence ratio.

It is defined as the ratio of the fuel-air ratio to the stoichiometric fuel-air ratio. Mathematically it can be expressed as

$$\phi = \frac{Y_{F,u}/Y_{O_2,u}}{(Y_{F,u}/Y_{O_2,u})_{st}}$$

(2.10)

The relationship between the mixture fraction and the equivalence ratio can be derived using the eq. 2.6, 2.7, 2.9 and 2.10

$$\phi = \frac{Z}{1-Z} \frac{(1-Z_{st})}{Z_{st}}$$

(2.11)

The above expression illustrates that the mixture fraction is related to the equivalence ratio.

In case of partially premixed combustion, the fuel stream is mixed with the air stream or vice versa before entering the burner domain. In this respect the conventional definition of the mixture fraction can not be used, due to the fact that the mixture fraction does not agree to its bound of 0 and 1, but may vary locally according to the partial premixing effects. A modified mixture fraction definition has
been proposed [90] to account for the partial premixing of the fuel/air streams. This modified mixture fraction can be expressed as follows:

\[ \xi = \frac{Z - Z_L}{Z_R - Z_L} \]  \hspace{1cm} (2.12)

where the \( Z_L \) and \( Z_R \) are the boundary values on the lean and the rich fuel side. \( Z \) represents the local value of the mixture fraction around the stoichiometry.

### 2.1.2 Reaction Progress Variable

The premixed turbulent combustion models assume the progress of the irreversible global chemical reaction through a normalized scalar which is known as reaction progress variable denoted as "c". This term accounts for the progress of the chemical reaction during combustion process in the premixed as well as in the partially premixed flames.

In closed systems, the reaction progress variable may be defined as a normalized function of temperature, since the temperature rises monotonically in closed adiabatic systems.

\[ c = \frac{(T_{\text{adia}} - T_{\text{min}})}{(T_{\text{max}} - T_{\text{min}})} \]  \hspace{1cm} (2.13)

where \( T_{\text{adia}} \) = adiabatic local temperature of partially burnt mixture in the combustion device

\( T_{\text{min}} = \) minimum temperature of the fresh reactants

\( T_{\text{adia}} = \) maximum temperature of fully burnt products

It can be observed from eq. 2.13 that the reaction progress variable takes the value of zero for the fresh reactants i.e., at the inlet temperature where no reaction has taken place and rises monotonically to unity in the fully burnt products i.e. at maximum adiabatic temperature attained at complete combustion.

In open non-adiabatic systems, the temperature rises monotonically to its maximum value and then decreases due to the heat losses to the environment. In that situation it is not possible to describe the progress of the reaction using the normalized temperature. The reaction progress variable in that case may be defined using the normalized mass fraction of the fuel in the burnt and the unburnt regime. Mathematically it can be expressed as:

\[ c = \frac{(Y_f - Y_{fr})}{(Y_{fp} - Y_{fr})} \]  \hspace{1cm} (2.14)

where \( Y_f \) = local fuel mass fraction concentration in the combustion device

\( Y_{fr} = \) fuel mass fraction concentration in the unburnt mixture

\( Y_{fp} = \) fuel mass fraction concentration in the fully burnt mixture
Types of Combustion and their Applicability

It may also be possible to use the stable end products of the combustion phenomenon for defining the reaction progress variables. In case of methane/air partially premixed combustion, the normalized mass fraction of CO$_2$ and H$_2$O can be used as shown below.

\[ c = \frac{Y_{CO_2}}{Y_{CO_2P}} \]  \hspace{1cm} (2.15)

or

\[ c = \frac{Y_{H_2O}}{Y_{H_2OP}} \]  \hspace{1cm} (2.16)

where $Y_{CO_2}$, $Y_{H_2O}$ = local CO$_2$ and H$_2$O conc. in the combustion devices

$Y_{CO_2P}$, $Y_{H_2OP}$ = CO$_2$ and H$_2$O conc. in the fully burnt mixture

It has been shown theoretically and experimentally that the reaction progress variable "c" calculated with different definitions for highly strained premixed flame tends to describe the flame quite well [20]. So for the calculation of the reaction progress variable, any normalised scalar definition may be used if the assumption of the highly strained flame is satisfied. However, the most relevant definition for the reaction progress variable is the one in which all the species are included [20].

2.2 Non-Premixed Combustion

Figure 2.1 shows the sketch used to define the non-premixed mode of the combustion.

![Figure 2.1 Sketch for defining the non-premixed mode of combustion](image)

As shown in the Figure 2.1, the non-premixed combustion involves the mixing of two streams i.e., fuel and air. The reaction takes place along the region where the stoichiometric conditions are fulfilled and the flame front is positioned along this stoichiometric mixture line. Since this combustion phenomenon requires the diffusion of the fuel and air species in the reaction zone, it is also known as diffusion combustion. The non-premixed combustion is mixing controlled. This type of combustion is a very stable combustion, however it involves very high peak temperatures thereby leading to very high NO$_x$ emissions.
This type of combustion mode appears in diesel engines, liquid fueled gas turbines, furnaces, steam boilers and fires.

2.2.1 Computational Models

The most widely used models for the nonpremixed combustion may be categorised into two types. The first category is that in which the time scale for the chemical reaction is very fast compared to the turbulence time scale. We define models lying in this type as fast chemistry models. The second type is that in which the time scale for the combustion reactions are of the order of turbulence time scale. We define these models as distributed reaction models. One example from each of these two types of models are discussed in the below.

The models based on the flamelet approach can be used for defining the flame structure of diffusion flame in case the reaction time scale is very short compared to the turbulent time scale. Therefore the turbulent flame may be regarded as an array of laminar flames "flamelets" embedded in the turbulent flow [46].

The laminar flamelet concept is designed for cases where the thickness of the flame front is much smaller than the smallest length scale of turbulence. This assumption requires that reactions get completed within the turn over of the smallest eddies. All chemical reactions and the heat release take place within the flamelets. With this assumption, the chemistry and the turbulence can be handled separately. The laminar flames can be computed separately using the detailed chemistry and the data can be stored in the form of flamelet libraries. All the species can be related to the conserved scalar, the mixture fraction, in case the assumption of equal diffusivity for all the species is applied.

The interaction of the flamelets with the turbulent flow field can be described with the help of a modelled scalar namely scalar dissipation rate $\chi$ or the strain rate $\alpha$. This modelled term accounts for the effect of the turbulence eddies on the structure of the flame front. The scalar dissipation rate in the laminar flamelets is related to the gradient of the mixture fraction in the physical space. The higher value of the $\chi$ can lead to the increase in the heat conduction from the reaction zone since the local gradient across the flame zone increases. If $\chi$ is increased beyond the critical value $\chi_c$, heat loss can no longer be balanced by heat produced due to chemical reaction and the flamelet is extinguished. In the turbulent flames this modelled term is a function of the inverse of the turbulence time scale ($k/\varepsilon$) and the variance of the mixture fraction ($\bar{Z}^2$), which accounts for the residence time of the turbulence eddies and the fluctuation of the mixture fraction about its stoichiometry in the physical domain.

Locally, The flamelets have a planar structure, since the flame thickness is sufficiently thin. So the flow in the reaction zone (flamelets) can be viewed as an opposed jet whose velocity field is given by a potential flow solution. The stagnation velocity gradient for such configurations can be defined as the strain rate. The strain rate is proportional to the inverse of the characteristic aerodynamic time. It has the similar interpretation as the scalar dissipation rate. As $\alpha$ is increased, the net rate of formation or consumption of the species and the heat release gradually increases. When the strain rate reaches some critical value, the flame is blown off implying limitation due to chemical kinetics.
Ferreira et al. [24] have shown numerically that it is appropriate to use the scalar dissipation rate as compared to the strain rate for discussing the impact of the turbulence on the flamelets in the nonpremixed turbulent flames. The strain rate does not consider the effect of the gradient of the mixture fraction, where as the scalar dissipation rate take these effects into account. However one can also use the strain rate for showing the effect of the turbulence on the laminar flame structure. Generally the strain rate is used in the premixed turbulent flames for describing the interaction of the turbulence and the chemistry in the flamelet model.

So in the standard models based on the flamelet approach, the coupling between the non-equilibrium chemistry and turbulence is achieved by the statistical description of these two parameters, the mixture fraction and the instantaneous scalar dissipation rate. In the steady flamelet model, the statistical independence of the mixture fraction and the scalar dissipation rate is assumed. The presumed shape of the probability density function (PDF) can be selected for the mixture fraction and the scalar dissipation rate for solving the mean species concentration in the turbulent flow field.

The regime of the validity of these models in non-premixed combustion holds true, only if certain conditions are satisfied. Generally these conditions can be expressed with non-dimensional parameters such as a turbulent Reynolds number, a Daemkoehlernumber and a Karlovitz number [75]. The steady and the transient laminar flamelet model [24] developed in our group is also based on the work of Peters et al. [75]. These models have been successfully investigated for the turbulent non-premixed jet flame and are able to predict the re-ignition and the extinction behaviour of the highly turbulent non-premixed jet flames [88] [27]. The TLFM model will be discussed later in detail for their predictions of the turbulent partially premixed flames.

The pdf (Probability Density Function) transport equation model shows promising results for the flame (e.g. Flame-E, F TNF Workshop) [68] having very high turbulence, where the flamelet assumption can not be applied. As mentioned before in the flamelet model a presumed pdf shape for the conserved scalars are used. However, in order to reproduce finite rate effects accurately, e.g. emissions, more independent scalars need to be incorporated in the characterization of the pdf. Moreover, in case of multi-scalar problems, the extension of presumed shape pdf methods is difficult. Such difficulties are averted through the use of pdf transport methods.

Pope [79] has done a pioneer work in the development of the Monte Carlo method for solving the composition joint PDF equations to predict the properties of the turbulent reactive flow fields. The transport equation for the density weighted scalar pdf is expressed as:
In the above equation the terms have the following significance:

Term I: Rate of change in physical space
Term II: Mean convection in physical space
Term III: Chemical source production in compositional space
Term IV: Turbulent transport in physical space
Term V: Molecular mixing in compositional space

The advantage of the PDF-transport equation model is that the chemical source term appears in closed form (cf. term III in the equation 2.17) and hence does not need to be modelled. But there are unclosed terms like turbulent transport and molecular mixing terms (cf. terms IV, V in the equation 2.17) due to the appearance of the conditional expectation, which have to be modelled in this method. However the computational requirement of these models increases linearly with the increase in the dimensionality of the PDF equations. So there is a restriction in the use of pdf equations of more scalar variables for the prediction of intermediate stable species in the turbulent non-premixed flames. Obieglo [70] has used the PDF transport equation model for the computation of the H2 jet flame and the piloted methane/air jet flame. He used Repro-modeling approach to incorporate detailed H2 chemistry in the form of spline functions. The functions were generated by fitting the simulation results obtained from the perfectly stirred reactor computations of a detailed hydrogen-oxygen mechanism. These functions contain the information of the intermediate species as the function of the time evolution of H2O and N2. The computational results in general show a good agreement with the experimental data.

Recently a CMC (Conditional Moment Closure) model, developed independently by Bilger and Klimenko [12], has been used for modelling the turbulent nonpremixed unsteady flames. In this method, the spatial and the temporal fluctuation of the unconditionally averaged quantities like species concentrations, can be obtained by integrating conditionally averaged values over the range of conserved scalar values after weighting with the local probability density function of the conserved scalar. In nonpremixed combustion, mixture fraction can be used as a conditioning variable for the conserved scalar. All the unconditioned variables can be related to this conditioned conserved scalar. The conditional average CMC equation of the mass fraction of the species \(i\) \(\langle Y_{i|\eta} \rangle\) can look like as eq. 2.18:
In the above equation \( v \) is velocity, \( W_i \) the rate of formation of the species \( i \) per unit mass of the mixture and \( N \) the scalar dissipation rate.

The scalar dissipation rate is defined as follows:

\[
N = D \nabla \xi \nabla \xi
\]  

where \( D \) is the molecular diffusivity, which is assumed equal for all the species.

Bilger et al. [13] have compared the CMC model and the steady flamelet model for the prediction of the turbulent hydrocarbon flames. They showed that the CMC model gives a better prediction as compared with the steady flamelet model predictions, after comparing the numerical results with the database generated by direct numerical simulation (DNS) for the non-premixed turbulent flames. It has been suggested that the applicability of the steady flamelet model is restricted to the condition, that the local reaction zone thickness is smaller than the turbulent dissipative length scale. In case this condition is not met, the turbulence inside the reaction zone may lead to substantial variation in the characteristic quantity, namely the scalar dissipation rate. In such situations the steady flamelet models can not be applied for determining the extinction/re-ignition behaviour of the turbulent non-premixed flames. The limitations of the steady flamelet model has been shown in the work of Ferreira [24].

Pitsch [77] has presented an unsteady flamelet model. The model describes the temporal variation of the flamelets that are transported downstream in the jet flames. He considered the convective transport along the stoichiometric mixture of the each flamelets introduced at the burner inlet. Ferreira developed unsteady flamelet libraries and then related these libraries in the turbulent flame using the modelled turbulence time scale. The flamelet model of Ferreira will be described later in the section of turbulent flames.

The above mentioned combustion models are widely used in the predictions of the minor species like CO, NO\(_x\), etc. in the turbulent non-premixed flames. There are limitations of the models for their use in the industrial applications, e.g. CMC and PDF-transport models are computationally very expensive models for computing complex industrial burners. The models based on the flamelet approach are used for modeling practical burners. However, these models may not give good predictions for the combustion phenomenon, where the turbulence length scales are of the order of the flamelet length scales. Moreover, the steady flamelet models are not capable of predicting the minor species like CO, due to the slow chemical reactions of the CO species. These models are continuously developed and optimized, such that they can be used as a regular CFD tool for the designing of the industrial burners.

It is worth mentioning here that the industrial combustion community often used generic combustion models, like Eddy Dissipation Model (EDM) [50], to study the interaction of the combustion and the turbulence in the burners operated in non-premixed modes. The mean reaction rate takes the minimum of three rates,
Types of Combustion and their Applicability

namely fuel, oxygen or products rates. Mathematically the mean reaction rate can be expressed as follows.

\[ \bar{\omega} = \rho A \frac{\varepsilon}{k} \min \left( \frac{Y_F}{Y_{O_2}}, \frac{B Y_P}{(1 + v)} \right) \]  

(2.20)

In the above expression A and B are model constants and v is the stoichiometric oxygen to fuel mass ratio.

This model is easy to use and also not computationally expensive. The model is based on the one step chemistry. It is not possible to predict the minor stable species with the usage of this model. However, the model is able to predict the heat release with good accuracy in certain industrial combustion applications [22, 76] and this helps in the development of the new burners.

2.3 Premixed Combustion

The sketch of the premixed combustion can be viewed in the Figure 2.2. In premixed combustion the fuel and air is mixed prior to the reaction and the mixture lies within the flammability limits. Normally these type of flames require some safety measures as the mixture can burn at places where it should not burn. If the mixture contains excess of air as usually needed for complete reaction, then this type of combustion helps in the reduction of the maximum temperature in the reaction zone, leading to lower emissions of nitrogen oxides. The premixed combustion is reaction controlled.

This type of combustion appears in spark ignition engines, gas-fuelled gas turbine engines and jet engine afterburners.

![Premixed Flame](image)

Figure 2.2 Sketch for defining the premixed mode of combustion

2.3.1 Computational Models

Similar to non-premixed combustion models, the models for the premixed combustion can also be divided into two types, namely fast chemistry models like the model based on the flamelet approach and slow chemistry models, where the detailed reaction rates are relevant and not the turbulence. One example of the fast chemistry model is the laminar flamelet model [19] and slow chemistry model, is
the pdf transport equation model. However, Schmid et al. [89] claimed that their model can be used in a wide range of Damköhler numbers.

In the flamelet model, it is assumed that the premixed flame front is a thin, continuous surface having the properties of the laminar flamelets. The reaction rate for such flamelets can be obtained through the detailed calculations of strained laminar flames defining the chemical composition in terms of a reaction progress variable. This scalar is already defined and explained in the previous section. The mean rate of the reaction progress variable is then computed through statistical averaging of the instantaneous term.

Bradley et al. [19] have introduced the simple flamelet model, in which the one-dimensional flamelets are described by the reaction progress variable. Under the assumption of the flamelet approach, the temperature and all the species concentrations are the function of the reaction progress variable. The mean values of the a scalar \( \phi \), namely, species and the temperature can then be calculated in the turbulent flame as

\[
\Phi = \int \Phi(c)P(c)dc
\]  

(2.21)

\( P(c) \) represent the probability density function of the reaction progress variable. They used a beta function for the reaction progress variable for the presented model. The shape of the beta function is determined by using the averaged first and second moments of the temperature. They applied this model in a jet stirred conical reactor. The model predictions of the stable species, such as CO are compared with the measured species concentrations. They found that the agreement between the experimental data and the computed results was good except in the region of high turbulent straining. The discussed model does not take into account the influence of curvature and the variation of the strain rate in time and space.

Benim et al. [7] have also used a laminar flamelet model based on the work of Bradley et al. for the simulation of the complex double cone burner of the ABB gas turbine. They used a double delta function for the reaction progress variable, due to the assumption of infinite fast chemistry. The reaction zone in premixed combustion is normally very thin and the chemical reactions are infinitely fast. So there is no probability of the reaction progress variable to have a value between 0 and 1. The strength and location of the delta functions are calculated by the mean value and the variance of the reaction progress variable. They have shown that the validity of the strained laminar flamelet model for the premixed combustion is beyond the limits as defined in the Borghi diagram [18], which is similar to the Peter's diagram [75] used for the nonpremixed combustion. The computational results showed a good agreement with the experimental data available for the gas turbine burner. Some differences between the experimental data and the computational results are observed by them and they suggested that these differences are due to the fact that in the flamelet model the variation of the strain rate was not taken into account. However the model is further developed to account for the effect of the variation in the strain rate [9] and gave a good prediction for the temperature and the species concentrations in the double cone burner of ABB gas turbine.
One of the earlier proposed model based on the assumption of infinitely fast chemistry is Eddy Break-up Model (EBU) of Mason and Spalding [53]. This model has been used extensively for understanding the effect of heat release in the industrial burners. The model proposed the following expression for the mean rate of production of the reaction progress variable

\[ \dot{\omega} = C_{EBU} \frac{\dot{\varepsilon}}{k} \sqrt{\gamma'^2} \]  

(2.22)

where \( C_{EBU} \) is a constant, \( k \) is the turbulence kinetic energy and \( \dot{\varepsilon} \) is the dissipation rate of the kinetic energy, \( \gamma'^2 \) is the variance of the mass fraction of the oxidant.

The limitation of EBU model is its inability to provide information about the intermediate species, which are required for predictions of the pollutants.

Schmid et al. [89] showed the capability of their model for a broader range of Damkoehler number from \( 0.2 < Da < 2.8 \). The Damkoehler number characterises the interaction between the large turbulent structures and the reaction zone.

\( Da >> 1 \) indicates that the chemistry is very fast in comparison to the turbulent motion and thereby no modification of the internal flame structure occurs. However, \( Da < 1 \) represents the condition where the turbulence intensities are very high such that combustion regime is distributed without having flamelet type structure. So this model can be classified as slow chemistry model owing to its ability to predict the distributed reaction regime. Schmid et al. have provided different formulations for the source term of the mean reaction rate for both fast as well as the slow chemistry phenomenon.

In the cases involving high turbulence intensities \( (Da, <<1) \), the reaction rate is limited by the chemical kinetics and expressed as:

\[ \dot{\omega} = 4.96 \frac{1}{\tau_{chem}} \bar{c}(1 - \bar{c})\rho_0 \sigma Y_{0, fue} \]  

(2.23)

In the cases lying within the flamelet regime \( (Da, >>1) \) the following expression for the source term of the reaction rate is suggested.

\[ \dot{\omega} = 4.96 \bar{c} \left( \frac{S_l}{\rho} + 1 \right) \frac{\varepsilon}{3k} (1 - \bar{c})\rho_0 \sigma Y_{0, fue} \]  

(2.24)

The eddy dissipation model as mentioned earlier can also be used for the numerical predictions of the premixed flames. Del Taglia et al. [21] have used this model for understanding the heat release phenomenon in a commercial household burner. They were able to utilize the capabilities of this generic model for the further development of this household burner.

Also PDF transport equation model can be used for investigating the premixed flames. In premixed combustion the transport equation of the pdf of reaction progress variable [80] is solved in a similar way as in non-premixed combustion
where the transport equation of the pdf of mixture fraction [70] is solved. There are unclosed terms involving turbulent micro-mixing and the reactions, which are to be modelled here in the pdf transport equation model for premixed combustion. At present the PDF transport equation models are not so often used in the combustion industry due to the high computational costs involved with it.

### 2.4 Partially Premixed Combustion

The phenomenon of partially premixed combustion is depicted in the Figure 2.3. In this type of combustion a fuel stream is mixed with air or an air stream mixed with fuel, such that the mixture still lies outside the flammability limits. Then the former stream is called a rich mixture and the latter a lean one. Both streams improve the starting conditions for the mixing process in the pre-flame zone. This helps in achieving a reduction in the maximum temperature while keeping the flame stable. So this type of flames can help in reducing the emissions. This behaviour of the partially premixed flames is going to be discussed in detail in the later sections.

![Figure 2.3 Sketch for defining the partially premixed mode of combustion](image)

Partially premixed combustion takes place in gas turbines with pre-vaporizers, direct injection automotive engines, gas fired furnaces, spray burning devices and also can be applied in bunsen burners.

#### 2.4.1 Computational Models

The developed models are not validated for predicting the reacting flow field in the turbulent partially premixed combustion modes in industrial burners, unlike the models that are available and validated for the accurate predictions of the reacting flow fields in non-premixed and premixed combustion modes. It may be due to fact that the usage of the partially premixed mode of combustion is relatively new in the industrial applications.

The partially premixed combustion can be obtained by the following two methods, namely
1. The lean and the rich mixtures are supplied as such to the burner, located on both side of the reaction zone and transferred to the combustion zone. This type of phenomenon is similar to the nonpremixed combustion. In the present thesis, such flames are known as partially premixed “diffusion” flames.

2. The partial premixing is produced in the premixed flames due to local quenching of the flame. The partially reacted species diffuse to the reaction zone in parallel, similar to the phenomenon of combustion of homogeneous mixture. These flames are known as partially reacted “premixed” flames in the present work.

The computational models for predicting the partially premixed “diffusion” flames should be developed by extending the existing models for non-premixed combustion to account for the non-homogeneous effects. The second approach is to extend the already validated premixed models to account for the partial premixing effects. These extended models should be suitable for the prediction of the partially reacted “premixed” and “diffusion” flames having combustion similar to the premixed combustion and non-premixed combustion.

Bilger [11] commented that the unsteady flamelet models and CMC model should be able to predict the flame structure for the turbulent partially premixed flames. He mentioned that the partially premixed flames should have similar flame structure as the non-premixed flames. Obieglo [70] used the pdf transport equation model for the prediction of the turbulent partially premixed piloted jet flame [68]. He was able to get good predictions of the species concentrations in the mixture fraction space. However, the model was unable to predict the correct length of the flame.

Rogg et al. [85] have proposed firstly the formulation for extending the laminar flamelet model of non-premixed combustion in order to account for the partially premixed phenomenon. In their model, they defined a term “premixedness parameter ζ” to characterize the degree to which the reactant streams of a flamelet are premixed.

Analog to the flamelet model for the diffusion flame, the proposed extended flamelet model for diffusion flames showing partially premixed structures suggests that any scalars, such as species concentrations or temperature, are functions of three variables, i.e. modified mixture fraction, scalar dissipation rate and the premixedness parameter. Mathematically, it can be expressed as

\[ \Phi = \Phi(\xi, \zeta, \chi_{st}) \]  

These variables are treated as random variables, such that a joint probability density function is required in order to calculate the species concentrations.

In the turbulent flow field, a relationship between the mean premixedness parameter and the mean mixture fraction is proposed.
where \( x \) denotes the distance measured from the burner exit, \( L \) the flame height, and \( \alpha \) the empirical constant. The above functional relationship is based on the assumption that the premixedness of the reactants increases with increasing distance away from the burner exit and also \( \zeta \) is 0 when mean mixture fraction attains its limiting value, i.e. 0 or 1.

They did computation of a high Reynold number diffusion flame by varying the empirical constant \( \alpha = 0, 5 \) and \( 10 \) and compared the computational results with the experimental data. They found that the computations carried out with \( \alpha = 10 \) for the extended flamelet model were able to give better predictions of the experimental flame structure as compared with the computations using lower values of \( \alpha \).

So these are the first computations that have used an extended flamelet model for the diffusion flames to show the existence of a partially premixed phenomenon in the high Reynolds number diffusion flame. Rogg et al. [85] suggested to use such type of formulation for the prediction of the turbulent partially premixed flames.

Peters et al. [60] have developed a model for the predictions of the partially premixed phenomenon in the turbulent jet flames. This model is based on the assumption of infinite fast chemistry. The flamelet models for premixed and non-premixed combustion are combined to predict the partially premixed flames. A two dimensional flamelet structure occurs at the intersection of the isolines describing the premixed reaction zone and the non-premixed reaction zone. In the non-premixed combustion the flamelets are located on the surface of the stoichiometric mixture fraction. In the laminar flamelet model for the premixed combustion, a new scalar, named \( G \), is introduced for describing the instantaneous flame contour as isoscalar surface of this scalar field. This isosurface propagates with the laminar burning velocity. This scalar function is identical in physics as the reaction progress variable. Moreover, this scalar plays a similar role for premixed flamelet combustion as the mixture fraction in the theory of non-premixed flamelet combustion. The transport equation for this scalar is

\[
\rho \left( \frac{\partial G}{\partial t} + \mathbf{v} \nabla G \right) = (\rho S_L) |\nabla G| \tag{2.27}
\]

where \( \rho S_L \) represents the constant mass burning velocity through a laminar plane steady flame. \( \mathbf{v} \) and \( \rho \) are the velocity and the density of the burnt gas. In a similar manner to the flamelet model for non-premixed combustion, the mean and the variance of the \( G \) scalar is derived. There are terms appearing during the derivation, which are to be closed by modeling.

The partially premixed flame structure propagates within a non-premixed mixing field following a surface of stoichiometric mixture fraction. The model showed good agreement with the experimental data for the liftoff heights of the turbulent diffusion jet flames. It is proved, that the partially premixed phenomenon occurs
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in the liftoff heights of the jet diffusion flames and this phenomenon causes the stabilization of the lifted jet nonpremixed flames [35, 108].

Favier et al. [25] proposed a model based on the two scalar level set approach for the modelling of the turbulent partially premixed combustion. This model is based on the flamelet approach and it combines the flamelet model for the premixed and the non premixed combustion. The G-scalar is used to identify the premixed flame front locations in the flow field, while the scalar $Z$ fixes the location of the diffusion flame in the flow field. This model is valid in the regime, where the chemical time and length scales are much smaller than the corresponding flow scales, so that the combustion takes place in thin flame zones. Outside these zones the flow is assumed to be in chemical equilibrium or inert. Ovink et al. [71] have used the model on these approaches and computed the lifted diffusion flames. They observed that the triple flames are considered as a key element in the stabilization mechanism of lifted flames.

Schmid et al. [89] also claims that their model is capable of predicting the partially premixed combustion. However, the application of this model for the prediction of the turbulent partially premixed flame is not known at the time of writing this thesis.

Veynante et al. [101] have proposed a combined flamelet model based on the concept of flame surface density. The model includes a balance equation for the flame area per unit volume. In the combined model, three such balance equations are solved for the local area of the diffusion flamelets, local area of the premixed flamelets and the local area of the nonreactive mixing zone separating the non-premixed reactants. The flame surface corresponds to the iso-surface of the mixture fraction in the non-premixed regime and to the iso-surface of the reaction-scalar (e.g. G-scalar from Peter's model) in the premixed combustion regime. It is not possible to predict the pollutants using this model. However, it can be used to predict the heat release and the burning speed of the partially premixed flames. This model has not been used for computing the industrial partially premixed burners.

Bajaj [2] has modified the strained laminar flamelet model [9] to account the effects of the partial premixing in the turbulent lean premixed combustion in the double cone burner of ABB company. The modified flamelet model was validated for three different cases for partially premixed combustion under lean operating conditions.

The transient flamelet model [24], which is developed in our group, is tested for the predictions of the turbulent partially premixed flames. The model is extended to account for the partial premixing effects in the diffusion flames. The capabilities and the limitations of this model would be discussed later in the thesis under the section of "turbulent partially premixed flames".

It has been shown that the flamelet approach is able to describe the turbulent partially premixed combustion mode and different forms of models are available based on this approach. So the above mentioned models can be used for the simulation of industrial burners operating on the partially premixed modes.
3 Laminar Partially Premixed Flames

This section gives an overview about the research that has been carried out, both experimentally and numerically, in understanding the structure of the laminar partially premixed flames and their potential to reduce the NO\textsubscript{x} emissions. Later our contributions will be discussed in detail in the field of investigation of the laminar partially premixed flames. We also investigated several reaction mechanism for methane-air combustion for their predictions of the laminar partially premixed flame structure and the NO\textsubscript{concentration}. The one dimensional code used for investigating the laminar counterflow burner is also described.

3.1 Literature Review

A number of research groups have contributed to the research of understanding the laminar partially premixed flames. Some of the group activities are briefly discussed in this section.

Puri et al. [91] have investigated laminar counterflow partially premixed flame both experimentally and numerically. They established a flame using $\phi_{\text{rich}} = 19$ and $\phi_{\text{lean}} = 0.5$. The partially premixed flame exhibits a diffusion and premixed flame structure. The numerical agreement of the major species like CH\textsubscript{4}, O\textsubscript{2}, N\textsubscript{2}, CO\textsubscript{2} and H\textsubscript{2}O are with the experimental data. However, the C\textsubscript{2}-species predictions are not good. They also showed that the partially premixed flame is less resistant to the strain rate near extinction as compared to the diffusion flame for the same velocity boundary conditions.

Gore, Puri and Takeno [96] have done both numerical and experimental investigations for quantifying the acetylene and ethylene mole fractions in methane/air partially premixed flames. They varied the fuel side equivalence ratio from 2.0 to 7.0 from one nozzle and kept pure air on the other nozzle for the counter flow burner. The numerical model used a detailed chemistry reaction mechanism containing 30 species and 278 reactions. The species were measured locally using gas chromatography. The numerical predictions were significantly higher than the measured values of the C\textsubscript{2}H\textsubscript{2} and C\textsubscript{2}H\textsubscript{4} species. They suggested that in order to find out this discrepancies, it is important to study the reaction rates of these species in the laminar partially premixed flames.

Puri et al. [113] investigated both computationally and experimentally two dimensional partially premixed methane-air flames. The Wolfhard Parker slot burner is used for the presented investigation. The rich fuel-air mixture is introduced from the inner slot and the pure air from the two outer slots. The rich mixture configuration is varied from equivalence ratio of 2, 2.5 and 3.0. As expected, they found the double structure of the flame with inner premixed reaction zone and outer non-premixed reaction zone. The intermediate products of H\textsubscript{2} and CO that are produced in the inner premixed flame, oxidized in the outer non-premixed flame. The agreement between the experimental data and the computational results are
good for the flame heights and the reaction zones. However, certain disagree¬
ment is attributed to the C₂ chemistry, which is not included in the mechanism
used.

Puri et al. [43] have investigated the burner which can represent the practical
burners operating under partially premixed mode. They studied the effects of the
flame chemistry on the flame structure by keeping a constant flow rate. The flame
structure of the laboratory flames, representing the practical burner, consists of a
region of rich combustion in which partial oxidation of the methane occurs to form
CO and H₂, followed by a nonpremixed flame in which complete oxidation of
these intermediate species occurs. They concluded that the flame structure of the
practical burners can be described by the application of the flamelet profiles.

The effect of the gravity on the flame structure of two dimensional partially pre¬
mixed flames have also been investigated by Puri et al. [112]. The purpose of the
study is to understand the structure of flames in micro-gravity situations, so that
some procedures can be developed to extinguish the fire that may outbreak in
space crafts. Normally the fires are of diffusion and partially premixed nature.
They observed that the rich premixed flame of the double flame structure, which
is established on the Wolfhard-Parker slot burner, burns more rapidly to produce
CO and H₂ in the gravity conditions. A double flame structure is observed both in
1-g and 0-g conditions. The spatial differences of the lean flame in 1-g and 0-g
are significant. However, the position of the rich flame does not vary in these two
conditions.

Puri et al. [107] have shown the double structure of the laminar partially premixed
coflow flame established on the Wolfhard-Parker slot burner using the method of
laser interferometric holography. This method can be employed to accurately de¬
termine the optical density of flames and thereafter, they used the temperature-
refractive index correlation for the determination of the temperature field in two di¬
mensional partially premixed flames.

Gore and Zhan [31] have investigated experimentally laminar partially premixed
coflow flames. They measured stable species, namely CH₄, O₂, N₂, CO, H₂, CO₂,
H₂O, NOₓ for 6 different partially premixed flames. They found non-monotonic be¬
haviour of the NOₓ emissions from φ = 4 till φ = 1.3. The observed behaviour can
not be explained either by the decrease in residence time, which should mono¬
tonically decrease the emission index, or by the increase in temperature, which
should monotonically increase the emission index. The definition of the emission
index is defined elsewhere [93] and also mentioned later in this thesis. Thus the
changes in prompt NO production rate caused by the changes in the intermediate
hydrocarbon chemistry might be responsible for the minimum of NOₓ emission in¬
dices observed between φ = 1.9 and 2.1. Also they measured the flame heights
and the radiative loss fractions. The flame height decreases with increasing level
of partial premixing. The radiative heat loss fractions show a small decrease with
increasing levels of partial premixing but reach a constant level. They observed a
double flame structure for φ ≤ 3.5. Our findings are similar to their observations
and will be discussed later.

Smooke et al. [8] studied both computational and experimentally the laminar par¬
tially premixed coflow flames. They investigated the effects of the partial premix-
Laminar Partially Premixed Flames

ing on the flame structure. They found the existence of the dual nature of the flame for a wide range of equivalence ratio in the main jet stream. The increase in the partial premixing results in strengthening the inner premixed flame front. This leads to less entrainment of the coflowing air to form the stoichiometric mixture, giving rise to increase in the peak centerline temperature for more partially premixed flames. They used GRI 2.11 reaction mechanism without the NOx chemistry for the computational work. They observed differences in the concentration of the C2H2 species after comparing the experimental data with the computational results. They presume that this difference may lie in the reaction scheme of the GRI 2.11 combustion mechanism.

Levinsky et al. [59] measured the NO concentration in laminar partially premixed methane-air coflow flames. They studied the impact of the upstream radiation heat losses on the emissions of the NO concentration. They found the NO emissions can be reduced by suppressing the Zeldovich mechanism of NO formation with the help of upstream heat losses and using rich mixture. They used equivalence ratio of 1.3 for main fuel jet and able to reduce the NOx emissions from 80 ppm to 40 ppm with their configuration, which can be used for the low NOx burner design.

Greenhalgh et al. [47] investigated experimentally the structure of the laminar counterflow partially premixed methane/air triple flame. They showed the merging limits of the premixed and the diffusion flame as a function of the strain rate and the equivalence ratio. The conditions defined the stability map for the laminar counter flowing methane-air triple flame. They also observed that the triple flame is more sensitive to rich flame stoichiometry than the lean flame stoichiometry, and it is also sensitive to axial strain rate. Our numerical findings are similar to the experimental findings of Greenhalgh et al. and it will be discussed in detail in the later section.

Kim et al. [42] did measurements of nitric oxide in laminar partially premixed ethane-air flames. They studied the effect of the premixing level at constant burner tube flow rate on the NO emissions. By keeping a constant total burner tube flow rate, they can eliminate the dependence of the overall fuel residence time on the changes in the premixing level. They also observed a dual structured partially premixed flame, consisting of an inner premixed flame and an outer nonpremixed flame in a coflow jet configuration. As expected, they observed the increase in the visible flame length with increase in the burner tube equivalence ratio, owing to the corresponding increase in the fuel flow rate. The flame shows a twin flame structure for \( \phi_{\text{rich}} < 2.2 \) and the flame exhibits a distinct change in structure with increasing \( \phi_{\text{rich}} \). They found that the NO emissions are minimum at \( \phi_{\text{rich}} = 2.2 \) showing the compromise of the NO production by the prompt mechanism at the premixed flame front and the thermal mechanism at the nonpremixed flame front. Similar findings are also published by Blum et al. [16], where they performed experiments on the modified ELCO household burner.

Mcenally et al. [56] have done an experimental study of non-fuel hydrocarbon concentrations (C\(_2\)H\(_2\)O, C\(_2\)H\(_4\)O, C\(_3\)H\(_4\)O, etc.) in coflowing partially premixed methane/air flames. They observed that partial premixing in general leads to lower pollutant emissions in coflow flames as compared with its diffusion counterparts. Also the flame length decreases with the increase in partial premixing.
Blum et al. [16] observed the similar behaviour of the flame length with partial premixing in their experiments.

Choi et al. [34] investigated the visible spectral characteristics of emissions from a partially premixed methane/air and a propane/air flame. The position of the CH and C₂ radicals are determined in the flames using image processing technique. They showed the intensities of the CH and C₂ radicals in methane/air partially premixed flame having equivalence ratio of the main jet stream as 3.3 and in propane/air flame with main jet equivalence ratio of 2.7.

Williams et al. [45] carried out computational and experimental tests on laminar partially premixed methanol-air flame. A reaction mechanism for the methanol-air combustion is presented in this paper. The computations are carried out using the FLAMEMASTER code. The predictions of the C₂ species, NOₓ, CH₄ are in good agreement with the experimental data for the methanol-air counterflow flame with the fuel side equivalence ratio of 2.4 and the strain rate of 50 [1/sec]. The flame structure consists of double flame structure, a premixed methanol/air flame and a diffusion flame of H₂/CO + air.

Takeno et al. [111] did a numerical study on NO emission characteristics of a partially premixed methane-air coflow flame. They used GRI 2.11 reaction mechanism for the computations. They compared the emissions of NO from 2D computational results of a coflow flame with the 1D computational results of the counterflow flame with the same fuel-air configuration. They found that in the coflow flame the NO production due to Fenimore mechanism is much larger than that in the 1D flame. Also the GRI mechanism overpredicts the NO formation through the Fenimore mechanism more than the Miller and Bowman NO mechanism. The contribution of the thermal mechanism is more or less the same for the M&B and GRI mechanism. This leads to a higher emission index for the whole range of equivalence ratios except the diffusion configuration with the use of GRI mechanism. The emissions are the same for the diffusion flame using both the GRI and the M&B mechanism.

Takeno et al. [61] studied numerically the NO emission characteristics of methane-air double flames. They discussed the negative production of the NO through prompt mechanism in the rich premixed flame. The effects of the velocity gradient and the equivalence ratio are discussed with respect to the NO emissions. They used C₂ reaction mechanism and NO mechanism compiled by Bowmann [57]. The similar behaviour for the double flame structure is also observed by us in the numerical studies, where we used the latest version of the detailed reaction mechanism [17]. We also investigated the reactions involved for the negative production of the NO in the rich premixed flames. These will be discussed in the section of the analysis of the reaction mechanism.

Subba et al. [30] have carried out experiments on a commercial household burner operated under partially premixed mode. They studied the effects of different designs of the burner exit port on the flame structure and the NO and CO emissions. They found that the NO emissions can be reduced by using a triangular burner exit port, however, at the expense of increase in CO emissions. This is one of the first papers, which describes the experiments on a commercial residential heating burner operating in partially premixed mode.
Recent predictions with the GRI-Mech 2.11 mechanism by Blevins et al. [15] have shown that at lower strain rates the rich premixed reaction zone in methane–air partially premixed double reaction zones stabilizes separately for $\phi \leq 2.5$. However, since it is difficult to establish an experimentally stable flame at very low strain rates, no experimental validations of this result are available. The change over from a single to a double reaction zone structure has been predicted to occur at a global strain rate of $150 \text{s}^{-1}$ for partially premixed flames established with a rich side equivalence ratio $\phi = 1.4–1.5$ by Smooke et al. [94].

Investigations of rich laminar partially premixed flames have been done by Williams et al. [94,105,106]. They have compared experimental data with simulations performed after implementing the CECR reaction mechanism for different equivalence ratios. These studies considered found that NO emissions can be reduced substantially by adding dilution agents (e.g., $\text{N}_2$, $\text{H}_2\text{O}$, and $\text{CO}_2$) in the fuel/air mixture. Laser induced fluorescence measurements [84] of NO in methane–air partially premixed counterflow flames have shown that NO emissions depend strongly upon the rate of the partial premixing of the fuel and air.

Recently Thomsen et al. [95] have carried out the LIF measurements and modelling of nitric oxide concentration in atmospheric counterflow premixed flames. They investigated three cases of flame configurations, namely $\phi = 0.65$, 0.9, 1.2. They implemented the GRI 2.11 reaction mechanism for the methane/air combustion. They found remarkable agreement between prediction and measurements for the NO concentrations under lean conditions. However, this agreement broke down in moderately rich flames both with respect to the peak NO concentrations and to the relative shape of the NO profiles. Our observations are also similar to their observations in the rich flames for the prediction of the NO concentrations. There are four major NO formation pathways, which are as follows - Zeldovich, $\text{N}_2\text{O}$ intermediate, prompt and NNH. Their findings suggests that the GRI mechanism predicts a greater contribution from the relatively unknown NNH pathway for the lean flames. The justification of such a large contribution of this pathway along with the Zeldovich pathway in the lean flames, is still an open question. They also commented that the good agreement of the NO predictions in the lean flames could just as easily be produced by conflicting inaccuracies in multiple pathways as by an overall accurate kinetic mechanism. This may be one reason for the poor performance of the mechanism in the rich flames.

It can be summarized that the previous investigations have considered the flame structure and extinction of laminar partially premixed flames, the effect of the rich mixture equivalence ratio on NO emission and the stabilization of the double flame structure with respect to the strain rate and the equivalence ratio. Most of the above numerical experiments are carried out using GRI 2.11 reaction mechanism for the methane-air combustion.

We investigated the partially premixed flames in order to contribute to the existing research carried out by the above mentioned groups. Our contributions will be discussed in the following sections.

Partially premixed flames have been shown to reduce NO$_x$ production substantially as compared to nonpremixed flames, provided the strain rate is appropriate for the required mixing. For instance, experiments on commercial household
burners operated under partially premixed conditions have shown that NO\textsubscript{x} emissions are reduced substantially through an appropriate amount of partial premixing. The reduction in NO\textsubscript{x} emissions is believed to occur due to the double reaction zone partially premixed flame structure at low strain rates [16]. We will discuss this assumption in detail in the next section. Earlier discussions of these flames in the literature (that are based on correlations of residence time, flame radiation, and the competing mechanisms of thermal and prompt NO\textsubscript{x} formation) have been unable to explain their NO\textsubscript{x} emissions across the entire range of burner equivalence ratios.

### 3.2 Technical Relevance of Partial Premixing

As mentioned before, Blum et al. [16] have measured NO\textsubscript{x} emissions in a commercial household burner, which is operated in the partially premixed mode. Figure 3.1 shows the NO\textsubscript{x} emission concentration ratio in the \( \phi_{\text{rich}} \) space for two values of \( \phi_{\text{lean}} = 0 \) and 0.1. The NO\textsubscript{x} emission concentration ratio is defined as

\[
CR_i = \frac{c_i(\Phi_B, \Phi_A, \Phi_D)}{c_i(\Phi_B = \infty, \Phi_A = 0, \Phi_D = 0.83)}
\]

where \( c_i \) indicates the measured concentration of a species \( i \) for a specific condition of equivalence ratios indicated within the parentheses. If \( CR_i \) equals unity, the concentration of this specific condition for the species \( i \) is the same as for the diffusion flame operated at the defined reference condition. For a \( CR_i > 1 \), the concentration is higher than for the reference condition and no reduction potential is present, whereas for a \( CR_i < 1 \) the opposite is true.

![Figure 3.1 NO\textsubscript{x} emission concentration ratio plotted versus the level of fuel tube equivalence ratio.](image-url)
Figure 3.2 Species mole fraction and temperature profiles in the physical space for $\phi_a=0.0$ and $\phi_b=1.7$

Experimentally it has been found that the NOx emissions start decreasing as equivalence ratio decreases from $\phi_{\text{rich}}=5.0$ towards the lower $\phi_{\text{rich}}$ values and minimum emissions are achieved in the region $\phi_{\text{rich}} \approx 1.7$.

Figure 3.3 Species mole fraction and temperature profiles in the physical space for $\phi_a=0.0$ and $\phi_b=2.7$
Early discussions in the literature could not explain the NO\textsubscript{x} behaviour at all values of burner tube equivalence ratios. Calculations of laminar partially premixed counter flow flames at lower strain rates showed a double flame structure [15] [109], which was also experimentally detected in laminar partially premixed co-flow flames [31] [41].

Figures 3.2 and 3.3 show temperature and species concentration profiles of the calculated laminar partially premixed methane/air flames for a global strain rate of K=20s\textsuperscript{-1}. The detailed configuration of the counter flow burner is described elsewhere [3]. GRI 3.0 reaction mechanism is implemented for the present simulations. A one-dimensional code, OPPDIF, is used for solving the steady state solution for axisymmetric flames established between two opposed flows [38] [48]. The code is described in the later section.

The profiles in above figures 3.2 are monitored for a rich mixture for the left inlet (\(\phi_B = 1.7\) for Fig. 3.2 and \(\phi_B = 2.7\) for Fig. 3.3) and pure air for the right one. The double flame structure with the rich premixed CH\textsubscript{4}/air flame on the left hand side and the non-premixed H\textsubscript{2}/CO/air flame on the right hand side is easily visible well for the lower equivalence ratio. For the other equivalence ratio a merged diffusion flame is observed. The second CH radical peak at the premixed CH\textsubscript{4}/air flame for the \(\phi_B = 1.7\) mixture is an indicator for the existence of a well separated rich flame. In the rich premixed flame, there is no oxygen available for the NO\textsubscript{x} production due to the prompt mechanism. This leads to lower total NO\textsubscript{x} emissions in laminar flames with twin flame structure as compared to the merged diffusion flames. A similar NO\textsubscript{x} trend appeared in the turbulent partially premixed flames, where the fuel tube equivalence ratio was varied. The similarity of those trend may explain the specific NO\textsubscript{x} behaviour in the turbulent partially premixed flames reported above (cf. Figure 3.1). Similar conclusion for the turbulent partially premixed flame is obtained if one consider the twin flame structure as a staged combustion where at first the fuel is burned in a rich premixed reaction zone at \(\phi = 1.7\) followed by a stoichiometric combustion.
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Figure 3.4 Species mole fraction and temperature profiles in the physical space for $\phi_a=0.1$ and $\phi_D=\infty$.

Figure 3.5 Species mole fraction and temperature profiles in the physical space for $\phi_a=0.3$ and $\phi_D=\infty$.

We have also performed numerical studies in order to investigate the NOx emissions in the laminar partially premixed flames by varying the equivalence ratio on the lean side (cf. Figures 3.4, 3.5). On the left inlet pure fuel is injected whereas on the right side a lean mixture enters ($\phi_a=0.1$ for Fig. 3.4 and $\phi_a=0.3$ for Fig. 3.5). We are unable to observe a double flame structure in both the figures. It may ap-
pear that the prompt mechanism for the NOx production does not change in these flames and no major differences are observed for the emissions due to partial premixing on the lean side. Such type of NOx behaviour is also observed in the turbulent partially premixed flames [16], where the fuel/air mixing is carried out on the lean side. This unique behaviour of the NOx emission is possibly an explanation of the NOx concentration behaviour in the above reported turbulent partially premixed flames.

Since experimental data for laminar partially premixed counter flow methane/air flames for lower equivalence ratios are missing, further work is needed to clarify the unique NOx trends for turbulent partially premixed co-flow flames.

We have also carried out numerical investigation for the above two cases of laminar counterflow burners in order to observe the effect of the strain rate on the NOx emissions.

Figure 3.6 shows the profile of the EINOx versus global strain rate for the two different cases of partial premixing (\(\phi_{\text{rich}} = 1.7\) and 2.7 and the \(\phi_{\text{lean}} = 0.0\)) for the counterflow burner geometry. The \(\phi_{\text{rich}} = 1.7\) case represents a double flame structure, whereas the \(\phi_{\text{rich}} = 2.7\) case gives rise to a single flame structure (cf. Figures 3.2, 3.3). The NOx index is defined as the NOx production rate per unit fuel consumption rate in the whole physical domain. The definition of NOx emission index (EINOx) is explained elsewhere [93]. This index is useful since it is independent of the burner geometry and could be utilized for the comparison work with other combustion configuration.

![Figure 3.6 Effect of the global strain rate on the EINOx emissions for two partial premixing configurations for the counterflow laminar flames.](image-url)
It can be observed that the NO\textsubscript{x} emissions for the case of $\phi_{\text{rich}} = 1.7$ is lower than the NO\textsubscript{x} emissions for $\phi_{\text{rich}} = 2.7$ case till a certain critical global strain rate. Afterwards the emission indices for $\phi_{\text{rich}} = 1.7$ case lies higher than the $\phi_{\text{rich}} = 2.7$ case. At lower strain rates a double flame structure is visible for the $\phi_{\text{rich}} = 1.7$ case and a single flame structure for $\phi_{\text{rich}} = 2.7$ case. The double zone structure comprises two separate reaction zones, one rich premixed and the other nonpremixed. The fuel/air mixture at the rich boundary lies just out of the flammability limit for the $\phi_{\text{rich}} = 1.7$ configuration, such that the separated flame establishes itself if sufficient energy and radical pool is available from the nonpremixed reaction zone. The position of the rich premixed reaction zone depends upon the energy flow from the corresponding nonpremixed zone. This double flame structure causes the reduction in the NO\textsubscript{x} emissions. At higher strain rate, there is not sufficient time available for the stabilization of the rich separated premixed flame and a single merged flame stabilizes. This in turn leads to higher NO\textsubscript{x} emissions due to excess oxygen available. The strain rate of $\phi_{\text{rich}} = 2.7$ case at higher strain rate is lower than $\phi_{\text{rich}} = 1.7$ case due to the lesser amount of oxygen available for the production of the NO emissions. In the merged flame, the NO reactions take place due to both prompt and the thermal NO mechanism.

Bachmeier et al. [1] have investigated experimentally in detail the formation of the nitric oxide in the rich hydrocarbon flames via prompt mechanism. In combustion rich regime ($\phi > 1.1$) the Zeldovich mechanism is not dominant since thermal mechanism is dominant in $T > 1800$ K. He observed that the prompt mechanism for the NO production starts at $\phi = 0.75$ for methane and $\phi = 0.6$ for the other hydrocarbon fuels. No prompt NO was found by them in their experiments for $\phi > 1.6$.

This justifies our assumption to use rich fuel configuration of $\phi = 1.7$. The partially premixed flame structure at this equivalence ratio can have a double flame structure at lower strain rate along with practically no prompt NO formation, leading to lower overall NO emissions as seen in the above ELCO burner measurements (cf. Figure 3.1).

We conclude that a particular range of values of partial premixing provides reduction in NO\textsubscript{x} emissions, and this aspect must be considered while designing partially premixed household burners.

### 3.3 Methodology

All the available experimental data are collated for rich laminar partially premixed counterflow methane–air flames and we have simulated the temperature and species concentrations in these flames by employing several chemical reaction mechanisms. The purpose of this work is to discuss the performance of these reaction mechanisms in the context of methane–air partially premixed flames. The influence of strain rate on NO formation and the flame structure will be considered. The conditions related to the existence of a double reaction zone structure and its potential to reduce NO emissions will also be discussed. We will also discuss the discrepancies on the rich side of the flame in the context of predictions by the various mechanisms.
3.3.1 Numerical Code - OPPDIF

The investigation of laminar partially premixed counterflow flames is performed using the one-dimensional OPPDIF code, which computes the steady state solution for axisymmetric flames established between two opposed flows [38] [48] along the centerline.

The following figure shows the sketch of the counterflow geometry used for the investigation of the laminar partially premixed flames. The distance between the two concentric counterflow jets is variable and the laminar flame stabilized near the stagnation plane. The location of the stagnation plane depends upon the momentum balance of the two streams. When the streams are partially premixed, two partially premixed flames may exist as shown in the Figure 3.7.

The code has been modified to account for radiation effects using a narrow band radiation model [33] [81]. Details regarding the code are as follows.

The assumption of linearly varying radial velocity in radial direction leads to a simplification in which the flow properties for potential flows are functions of axial distance only, thereby reducing the two dimensional flow to one dimension model. This can be achieved by defining a stream function: \( y(x,r) = r^2 U(x) \), which satisfies the mass continuity equation.

Figure 3.7 Sketch of the Counterflow burner configuration

Using the definition of the stream function, it is possible to define the following two parameters:
Then the equations are reduced to the following set of one dimension equations, such that the continuity equation reduces to:

\[ G(x) = \frac{dF(x)}{dx} \]  

(3.3)

And the momentum equation:

\[
H - 2 \frac{d}{dx} \left( \frac{FG}{\rho} \right) + 3G^2 \frac{d}{dx} \left( \mu \frac{d}{dx} \left( \frac{G}{\rho} \right) \right) = 0
\]

(3.4)

where

\[ H = \frac{1}{r} \frac{\partial p}{\partial r} = \text{const.} \]

(3.5)

The energy equation:

\[
\rho u \frac{dT}{dx} - \frac{1}{c_p} \left( \lambda \frac{dT}{dx} \right) + \frac{P}{c_p} \sum_k c_{\rho k} Y_k V_k \frac{dT}{dx} + \frac{1}{c_p} \sum_k h_k \dot{\omega}_k = 0
\]

(3.6)

The species equation:

\[
\rho u \frac{dY_k}{dx} + \frac{d}{dx} \left( \rho Y_k V_k \right) - \dot{\omega}_k W_k = 0 \quad k = 1, K
\]

(3.7)

In the above set of equations \( c_p \) is the specific heat, \( h_k \) are the species molar enthalpy, \( W_k \) are the species molar mass and \( \omega_k \) are the chemical production rates by chemical reaction. \( \mu \) is the dynamic viscosity, \( \lambda \) is thermal conductivity, \( p \) is the density of the mixture and \( V_k \) is the diffusion velocity, which can be defined using the following two formulation approach:

- Multi-component formulation

\[
V_k = \frac{1}{X_k} \sum_{j=1}^{K} W_{kj} D_{ij} \frac{dX_j}{dx} + \frac{1}{\rho Y_k T} \frac{dT}{dx}
\]

(3.8)
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- Mixture-averaged formulation

\[
V_k = - \frac{1}{X_k} D_{km} \frac{dX_k}{dx} - \frac{D_k^T}{\rho Y_k} \frac{1}{T} \frac{dT}{dx}
\]  

where

\[
D_{km} = \frac{1 - Y}{\sum_{j=1}^{k} X_j / D_{jk}}
\]

where \(X_k\) are the mole fractions, \(D_{kj}\) are the ordinary multi-component diffusion coefficients, and \(D_k^T\) are the thermal diffusion coefficients.

The input conditions for the OPPDIF code is mixture composition (mole fraction), temperature (Kelvin) and the inlet velocities (cm/sec) at the nozzles exits.

Then the boundary conditions may be defined for the present case as:

\[
x = 0: \quad F = \rho_{RM} u_{RM}/2, \quad G = 0, \quad T = T_{RM}, \quad \rho u Y_k + \rho Y_k V_k = (\rho u Y_k)_{RM}
\]

\[
x = L: \quad F = \rho_{LM} u_{LM}/2, \quad G = 0, \quad T = T_{LM}, \quad \rho u Y_k + \rho Y_k V_k = (\rho u Y_k)_{LM}
\]

Therefore the inflow boundary condition specifies the total mass flux, including the diffusion and convection.

The Chemkin [39] [40] package provides the reaction rates, thermodynamic properties and the transport properties. So using the above stated differential equations and the appropriate boundary conditions, the boundary value problem can be solved for the dependent variables (\(F, G, H, T, Y_k\)).

A Central Differencing Scheme is used for the diffusive terms and an Upwind Differencing Scheme is used for the convective terms in the set of differential equations to be solved. The solver named “twopnt boundary value” [32] solves the discretized equations. The Twopnt uses Newton's method of iteration and the time integration method to calculate the steady state solution. So in order to attempt for the steady state solution the solver may go back and forth between the Time integration and Newton's method several times before converging. So this one-dimensional model computes the steady state solution for an axisymmetric partially premixed flame.

In the modified version of the OPPDIF code, the energy equation is extended to take into account the effect of the radiation [33].

The steady state solutions for axisymmetric laminar counterflow partially premixed flames for two different counterflow separation distances, i.e. 10 mm and
Laminar Partially Premixed Flames

18 mm are presented. The first configuration is utilized to understand the influence of strain rate on NOx emissions and the flame structure. The second corresponds to the boundary conditions for the experiments of Williams et al. [106]. In this case the reduction of the NOx emission index is discussed in context of the aforementioned double reaction zone structure. Additional experimental data is also considered [94] [105] [84].

3.3.2 Chemical mechanisms

Several chemical reaction mechanisms to predict the structures of atmospheric methane–air flames have been implemented. These mechanisms are described as below:

GRI Mechanisms (Frenklach et al. [17]):

GRI (Gas Research Institute) - Mech is an optimized detailed chemical reaction mechanism capable of the representation of natural gas flames and ignition. In the present study the following two latest version of the GRI-Mech has been implemented.

• Version 2.11, which contains 49 species and 279 reversible reactions.
• Version 3.0, which contains 53 species and 325 reversible reactions.

CECR Mechanism (Li and Williams [104]):

This mechanism is developed as part of the research at CECR, University of California, San Diego, USA. In our group we have implemented the latest version of the CECR mechanism (Version 4.0, 1999) which contains 54 species and 196 reversible reactions.

CEC Mechanism (Warnatz [103]):

Warnatz et al. from University of Heidelberg have developed the CEC reaction mechanism for the methane-air combustion. Here the 1997 version is implemented in the carried out computations. This version contains 53 species and 301 reversible reactions.

All these mechanisms are compatible with the CHEMKIN software and are available on the webpage of our combustion group.

3.3.3 Method of Analysis

An analysis of the reaction mechanisms is performed by applying the KINALC code [99] to the OPPDIF solutions. KINALC is built as an extension to the Sandia Laboratories CHEMKIN [40] package, and can extract information from the sensitivity matrices provided by simulation programs such as OPPDIF. KINALC can process the information about the concentration sensitivity in the following different ways [98]:

1. The program can extract the important information from the sensitivity matrices produced by the simulation programs. KINALC normalizes the sensitivity ma-
trices using the equation 3.11. It produces ordered lists of sensitivities of selected species and temperature at selected reaction times or distances.

\[
S_{ij} = \frac{k_i \partial c_i}{c_i \partial k_j}
\]  

(3.11)

In the above expression \(S_{ij}\) denotes the normalized sensitivity of the concentration \(c_i\) of \(i^{th}\) species to the reaction constant \(k_j\) for \(j^{th}\) reaction. (This expression is readily understood in context of the analogous sensitivity for the laminar flame speed)

\[
S = \frac{k_i \partial \nu}{\nu \partial k_j}
\]  

(3.12)

where \(\nu\) denotes the laminar flame speed.

2. KINALC can calculate the overall sensitivity using the equation 3.13. The user has to provide the information about the species involved in the function and the program provides ordered overall sensitivities at any time or distance.

\[
B_j = \sum (S_{ij})
\]  

(3.13)

3. This program can be utilized in carrying out principal component analysis of the concentration sensitivity matrix. The principal component analysis is a mathematical tool that assesses the effect of simultaneously changing parameters (e.g. temperature, distance, time) on several outputs (reaction rates, concentrations) of a model. The objective function of the principal component analysis is the square of the normalized deviation of concentrations, summed over all concentrations and for all time points. Mathematically the function is expressed as shown in equation 3.14. So the input to this function is the list of species, and the time or distance interval to be investigated. Also sensitivity analysis can be applied in the reduction of mechanisms and principal component analysis has been applied several times to the reduction of a mechanism [97].

\[
\alpha = \sum_{h=1}^{1} \sum_{i=1}^{n} \left( \frac{\Delta c_i(t_h)}{c_i(t_h)} \right)
\]  

(3.14)

where \(\Delta c_i(t_h)\) is the deviation of concentration at time \(t_h\) as result of parameter change \(\Delta \alpha\), where \(\Delta \alpha = \alpha - \alpha^0\), \(\alpha^0 = \ln(k_j)\).

4. A reaction is redundant if its sensitivity is small with respect to each species of the reaction system at all times in the interval considered. On the other hand, the reaction with high sensitivity is considered to be a rate limiting reaction, such that an increase in its rate coefficient causes a significant change in the overall reaction rate. KINALC can determine the rate limiting steps.
KINALC can also be used for determining the rate of production analysis and calculation of the fluxes of elements from species to species and contribution of each reaction to these fluxes.

In this investigation the program is used to determine the sensitivity of various species on the rich side of partially premixed flames. In this manner we are able to identify potentially problematic reactions, i.e., those that might cause incorrect predictions of the double reaction zone structure for those flames. This program is also utilized to further understand the NO formation pathways.

### 3.3.4 Definition of the strain rate concept

Flame stretch or strain rate is the relevant concept in turbulent combustion. In the premixed turbulent flows the flame front gets curved due to the impact of the turbulence on the flame speed. The curved flame front changes its surface while propagating, thereby effecting the rate of combustion. The relative rate of change of flame surface is called flame stretch and is expressed mathematically as [28]:

$$s = \frac{1}{A} \frac{dA}{dt} = \frac{d(\ln A)}{dt} \quad [s] \quad (3.15)$$

If the flame stretches beyond a certain limit, then the heat loss from the curved flame is so big that the flame can locally extinct. In the laminar diffusion flames the stretch or strain rate is defined by the velocity gradient along the axis of the flame.

The different forms of the definition of the strain rate are briefly discussed, since these terms will be required to interpret the results in the next section. The characteristic strain rate for a laminar counterflow flame (Global strain rate) [82] is defined as

$$K = \frac{2(-u_o)}{L} \left[ 1 + \frac{u_F}{(-u_o)\sqrt{\rho_F/\rho_o}} \right] \quad (3.16)$$

where \( L \) denotes the distance between the nozzles, \( u_o \) and \( u_F \) are the axial velocities on the air and fuel sides, respectively, and \( \rho_o, \rho_F \) denote the respective densities. When the computational results are compared with the measurements of Williams et al. [105, 106], the air side strain rate is employed to be consistent. This strain rate is the gradient of the axial velocity on the air side [105], namely,

$$a = \frac{du_{air}}{dx} \quad (3.17)$$
3.4 Results and discussion

3.4.1 Effect of partial premixing on the flame extinction

The Table 3.1 shows the local extinction maximum temperature and the global strain rate for different boundary configuration for air/fuel mixture, which have been simulated. The velocities of both the rich and the lean stream is kept same. The definition of the global strain rate is already defined earlier in the previous section.

It can be observed that the partial premixing does not have any substantial effect on the local extinction temperature (ΔT for the extreme case is ~ 40°K) but it has a big impact on the global strain rate at extinction. The flame extinguishes at a lower strain rate for extreme case with ϕ_{rich} = ∞. It is observed for a particular ϕ_{lean} value (ϕ_{lean} = 0), that the extinction strain rate increases with the increase in the partial premixing on the fuel side. The radical pool produced at the oxidizer side of the diffusion flame is consumed by the premixed flame on the rich fuel side. The increase in the partial premixing leads to the increase in the availability of the oxidizer for the complete combustion of the intermediate species. However in case of ϕ_{lean} = 0.1, the extinction strain rate increases with increase in the partial premixing till a certain configuration (ϕ_{rich} = 1.7) and then the global strain rate at the extinction decreases (cf. column ϕ_{rich} = 1.4) with the increase in the partial premixing. The general trend that is observed from the present numerical investigation shows that the partially premixed flame is more resistant to the strain rate near extinction as compared to its diffusion counterpart with increase in partial premixing.

<table>
<thead>
<tr>
<th>ϕ_{rich}</th>
<th>T_{max} [°K], K[1/sec]</th>
<th>T_{max} [°K], K[1/sec]</th>
<th>T_{max} [°K], K[1/sec]</th>
<th>T_{max} [°K], K[1/sec]</th>
<th>T_{max} [°K], K[1/sec]</th>
<th>T_{max} [°K], K[1/sec]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>1767, 844</td>
<td>1758, 842</td>
<td>1747, 772</td>
<td>1749, 704</td>
<td>1744, 640</td>
<td>1784, 468</td>
</tr>
<tr>
<td>0.1</td>
<td>1751, 868</td>
<td>1748, 901</td>
<td>1746, 807</td>
<td>1742, 762</td>
<td>1752, 694</td>
<td>1772, 503</td>
</tr>
</tbody>
</table>

Table 3.1: Maximum flame temperature and the global strain rate at different fuel/air mixture near local extinction limits

3.4.2 Effect of strain rate on NOx Emissions

The effect of the global strain rate on NOx production in laminar partially premixed flames is now discussed. The counterflow inlet velocities have been varied to change the global strain rate. Figure 3.2 presents the spatial distribution of NOx production for a non-radiating flame established at a stoichiometry corresponding to ϕ_{lean} = 0.1 and ϕ_{rich} = 2.8 and a counterflow separation distance of 10 mm. The predictions correspond to the GRI-Mech 2.11 reaction mechanism. For this case, we observe no significant differences in the flame structure for predictions ob-
tained with the GRI 2.11 and the GRI 3.0 mechanisms. We observe that the peak NO\textsubscript{x} production first increases with an increase in the strain rate until a critical strain rate (K = 390 s\textsuperscript{-1}), and decreases thereafter until the flame finally extinguishes.

The decrease in NO\textsubscript{x} production is due to the larger heat dissipation from the reaction zones established in the higher strain rate flames (above K = 390 s\textsuperscript{-1}), which lowers the peak temperature and, therefore, also lowers the formation of radicals that lead to NO\textsubscript{x} formation. At lower strain rates, the rich premixed flame can not establish itself for the present configuration, as the rich mixture lies outside the flammability limits and is too rich to burn itself. However the reaction zone is broader as compared to the reaction zone thickness for the flames established at higher strain rates. Both the prompt as well as the thermal mechanisms play an important role for the NO formation in the present laminar partially premixed flame.

The peak flame temperature decreases continuously as the strain rate is increased, but the NO\textsubscript{x} production does not follow this behaviour. The reactions involved with NO formation are the same for these flames. However, the reaction rates vary, which influences NO\textsubscript{x} production with respect to varying strain rate. The peak NO production increases with an increase in the strain rate until K = 195 s\textsuperscript{-1} due to the increase in the key reaction rate in the thermal mechanism, namely.

![Figure 3.8 NO\textsubscript{x} production rate profiles in the physical space for the configuration \(\phi_{\text{lean}} = 0.1\) and \(\phi_{\text{rich}} = 2.8\) showing the effect of the strain rate on the NO\textsubscript{x} production.]

The peak flame temperature decreases continuously as the strain rate is increased, but the NO\textsubscript{x} production does not follow this behaviour. The reactions involved with NO formation are the same for these flames. However, the reaction rates vary, which influences NO\textsubscript{x} production with respect to varying strain rate. The peak NO production increases with an increase in the strain rate until K = 195 s\textsuperscript{-1} due to the increase in the key reaction rate in the thermal mechanism, namely.
N + OH → NO + H (cf. Figures 3.9, 3.10, 3.11, 3.12), and then decreases as the strain rate is further increased. There is a better mixing of the radicals involved in this reaction as the strain rate increases. However, the NO consumption rate due to the reaction \( \text{CH}_2 + \text{NO} \rightarrow \text{H} + \text{HNCO} \) (cf. Figure 3.11) reaches a maximum value at \( K = 195 \, \text{s}^{-1} \) and this reaction rate decreases with further increase in the strain rate (cf. Figure 3.12). It is clear in this context that modifying the strain rate helps to reduce the \( \text{NO}_x \) emissions for a preset fuel/air boundary condition.

![Diagram](image_url)

**Figure 3.9** Contribution of the reactions for the \( \text{NO}_x \) production rate profiles in the physical space for the configuration \( \phi_{\text{lean}} = 0.1 \) and \( \phi_{\text{rich}} = 2.8 \) for the global strain rate of 78 [1/sec].
Figure 3.10 Contribution of the reactions for the NO\textsubscript{x} production rate profiles in the physical space for the configuration $\phi_{\text{lean}} = 0.1$ and $\phi_{\text{rich}} = 2.8$ for the global strain rate of 195 [1/sec].

Figure 3.11 Contribution of the reactions for the NO\textsubscript{x} production rate profiles in the physical space for the configuration $\phi_{\text{lean}} = 0.1$ and $\phi_{\text{rich}} = 2.8$ for the global strain rate of 390 [1/sec].
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1. \[ K = 585 \text{ [1/sec]} \]

2. \[ N + OH \leftrightarrow NO + H \]

3. \[ H_2O \leftrightarrow H + NO + M \]

4. \[ HNO \leftrightarrow H_2 + NO \]

5. \[ CH + NO \leftrightarrow HCN + O \]

6. \[ CH + NO \leftrightarrow N + HCO \]

Figure 3.11 Contribution of the reactions for the NOx production rate profiles in the physical space for the configuration \( \phi_{\text{lean}} = 0.1 \) and \( \phi_{\text{rich}} = 2.8 \) for the global strain rate of 585 [1/sec].

3.4.3 Comparison of the experimental data and the computational results - Flame Structure

We now compare the experimental data of Ref. 106 with predictions, mainly for temperature profiles and major species by three different reaction mechanisms. The results pertain to different fuel/air configurations for the counterflow geometry. Partial premixing with air is implemented on the fuel side and air alone is provided through the opposing stream. The air side strain rate is held constant at 50 s^{-1}. Figure 3.13 presents a comparison of the temperature profile for the \( \phi_{\text{rich}} = 1.5 \) case. The predicted maximum temperature is higher than the measured value, which is probably due to the fact that we have not accounted for thermal radiation in these simulations. The lean side flame structure is well predicted, but discrepancies are observed on the rich side.

The flame structure predicted by the GRI 2.11, GRI 3.0 and CEC mechanisms is similar. The CECR mechanism more accurately predicts the position of the rich premixed reaction zone, which has been also observed by Williams et al. [106]. A double reaction zone structure is simulated by the CECR mechanism, but the other mechanisms are unable to predict the correct position of the rich premixed reaction zone and such a structure. Blevins et al. [15] also observed numerically a double flame structure at lower strain rates.

The double zone structure comprises two separate reaction zones, one rich premixed and the other nonpremixed. Since the fuel/air mixture at the rich boundary lies within the flammability limit for the present configuration, this mixture is self-ignitable if sufficient energy is available from the nonpremixed reaction zone. The
position of the rich premixed reaction zone depends upon the energy flow from the corresponding nonpremixed zone. Reactants are consumed differently in the two zones. In the rich premixed zone, the methane/air mixture reacts to produce the stable species CO and H2. These intermediates are convected towards the nonpremixed zone and oxidized by the oxygen supplied from the lean side. The profiles of the major species for this case of $\phi_{\text{rich}} = 1.5$ are discussed in the next paragraphs.

![Image](image_url)

**Figure 3.13** Comparison of the experimental data and the computational results in the physical space for the temperature profiles for the configuration $\phi_{\text{lean}} = 0.0$ and $\phi_{\text{rich}} = 1.5$ (Li & Williams Flame).

Figure 3.14 shows the comparison of the profiles for the reactants (CH4, O2, N2) for $\phi_{\text{rich}} = 1.5$. The figure shows the numerical results obtained by using GRI 2.11 and GRI 3.0 reaction mechanisms. The N2 profile for the computations shows quite a good agreement with the experimental data. The consumption rate for methane and air show a good qualitative agreement with the experimental data, although a shift of the slopes toward the lean side can be observed in the computations. This may be due to the fact that the measured flame is broader, which shows that the heat diffuses faster towards the rich side causing the reaction to start 4 mm downstream from the fuel nozzle exit in experiment. The computed results are unable to predict this location of the CH4/O2 reaction. This may be due to the underprediction of the laminar flame speed.
Figure 3.14 Comparison of the experimental data and the computational results in the physical space for the reactant profiles (CH₄, O₂, N₂) for the configuration $\phi_{\text{lean}} = 0.0$ and $\phi_{\text{rich}} = 1.5$.

Figure 3.15 Comparison of the experimental data and the computational results for the products profiles (H₂, CO, CO₂) in the physical space for the configuration $\phi_{\text{lean}} = 0.0$ and $\phi_{\text{rich}} = 1.5$. 
Figure 3.15 shows the major product species (CO, H₂, CO₂) for φ_{rich} = 1.5 case. It can be observed that the calculated CO₂ profile shows a good agreement with the experimental data, except on the rich side. The observed differences are expected as the reaction of methane/air rich mixture starts at that position (cf. CH₄, O₂ profiles in Figure 3.14). The position of the rich flame depends upon the diffusive transport of the heat from the diffusion flame. The CO profiles also show good agreement with the experimental data except on the rich side. The H₂ computed results exhibit a large difference with the experimental data. A possible explanation may be attributed to the experimental uncertainties involved in measuring the H₂ species concentration [106].

Figure 3.16 Comparison of the experimental data and the computational results for the products profiles (C₂H₆) in the physical space for the configuration φ_{lean} = 0.0 and φ_{rich} = 1.5.

Figure 3.16 shows the C₂H₆ concentration profiles for the case φ_{rich} = 1.5. In this figure, the C₂H₆ concentration profiles obtained with GRI 2.11, GRI 3.0, CECR and CEC reaction mechanism are shown. It can be observed that all the tested mechanisms are unable to capture the position of the peak concentration. Moreover, the position of the peak concentration is more or less same for GRI 3.0, GRI 2.11 and CEC reaction mechanism. The CECR reaction mechanism predicts better position of the peak concentration as compared with the other mechanisms. However, it overpredicts the C₂H₆ concentration. The peak C₂H₆ concentration is overpredicted by all the tested reaction mechanisms. The profiles of the concentration of C₂H₆ is similar for both the GRI 3.0 and CEC reaction mechanism. GRI 2.11 predictions of the concentration profile are higher than that of GRI 3.0 reac-
tion mechanism. CECR mechanism predictions of the peak concentration is higher as compared with the GRI 3.0 and CEC reaction mechanism predictions, but lower than the predictions of the GRI 2.11 reaction mechanism. Williams et al. [106] have used their mechanism (CECR) for the similar numerical calculations and our results are similar to their numerical results. It can be stated that none of the mechanism is able to capture the C$_2$H$_6$ concentration. The C$_2$ chemistry plays an important role in the NO$_x$ mechanism on the rich mixture side.

The double zone structure explains the predicted reduction in the NO emission as compared to a comparable nonpremixed flame. In the rich premixed region little oxygen is available for NO production through the prompt mechanism. This leads to lower total NO$_x$ emissions from laminar flames that have a double reaction zone structure as compared with nonpremixed flames.

We have simulated plug flow conditions at the nozzle inlet, but the experimental boundary conditions probably lie between the conditions with or without the radial velocity components. This difference between the simulations and experiments may help to explain the small discrepancies between the rich side experimental data with results obtained by using the CECR mechanism.

Figure 3.17 presents a comparison of the predicted temperature profiles using the CECR and GRI 3.0 reaction mechanisms after implementing the radiation model with experimental data for the $\phi_{\text{rich}} = 1.5$ flame. Both mechanisms are able to pre-
dict the peak temperature with acceptable accuracy. However, the predictions differ regarding the position of the rich premixed reaction zone, and its attachment to the nonpremixed reaction zone. As mentioned above, the position of the rich premixed reaction zone depends upon the energy transfer from the nonpremixed zone. The heat generated from the premixed flame plays a role in determining the position of the rich flame. This can be determined by the speed of the reaction at this position. The position of the rich zone shifts toward the nonpremixed zone in the simulations that account for radiation (cf. Figure 3.13). The reason for this is that there is a smaller amount of heat transfer in this case from the nonpremixed to the rich premixed reaction zone, which is responsible for stabilizing the rich zone.

Figure 3.18 presents temperature profiles for the counterflow flame considered by Smooke et al. [94] at a global strain rate of 150 s\(^{-1}\) and a 1.27 cm separation distance. The fuel side equivalence ratio is 1.4. The flame structure is measured with laser diagnostics and a double reaction zone structure is observed, both numerically and in the experiments by the ref. [94]. They used a methane oxidation mechanism including NO\(_x\) chemistry, which consists of 43 chemical species and 158 reversible chemical reactions [92]. They compared the experimental data with the computational results and observed some differences in the temperature gradient of the rich premixed region. However, they confirmed the appearance of the double reaction zone structure by the Raman measurements.
We also observed the double reaction zone structure for the present configuration using different reaction mechanisms. The various mechanisms differ with respect to the rich side flame position, but behave similarly on the lean side. The CECR mechanism predicts a broader flame in comparison with the other mechanisms. Again, the reaction mechanisms differ with respect to the prediction of the position of the rich premixed reaction zone and its synergistic coupling with the nonpremixed zone. We observed a similar behaviour of the mechanisms for the numerical prediction of the flame structure on the rich and the lean side of the Li and Williams flame (cf. Figure 3.13, 3.17). The correct prediction of the position of the rich premixed reaction zone in the context of a double reaction zone structure is important for an accurate prediction of NO emissions.

Figure 3.19 presents a comparison of the temperature profiles predicted (accounting for thermal radiation) using the various reaction mechanisms with the experimental data of Laurendeau et al. [84]. The position of the rich premixed reaction zone is predicted well by the CECR mechanism, but the other mechanisms are somewhat deficient in this regard. The CECR mechanism predicts a broader flame (in accord with the measurements) than the other mechanisms. However, all of the mechanisms are unable to correctly predict the peak temperature. The computed peak temperature is about 200-300K lower than the measured peak temperature. This disagreement may be due to the experimental technique [84], in which an assumption of constant emissivity with respect to wavelength and temperature was employed for measuring emission from SiC fibers. Not much is
known about physical or chemical changes that might occur to the fiber material at temperatures approaching 2000 K. The simulations obtained with the GRI mechanism are in correspondence with the predictions of Ref. 84, which employed the same mechanism.

Figure 3.20 presents temperature profiles for the $\phi_{\text{rich}} = 2.5$ flame considered in Ref. 106. The simulations consider a non-radiating flame, but show good agreement with the measured flame thickness. The predicted peak temperature is higher than in the measurement, which is to be expected since radiation effects are neglected. The various reaction mechanisms exhibit similar behaviour over the entire flame, both on the rich and the lean sides. In the present configuration, the fuel/air mixture at the fuel nozzle inlet is very rich and does not lie within the flammability limits. The rich premixed reaction zone does not establish itself, even if sufficient energy is available from the nonpremixed zone. The flame structure for such flames consists of a merged single reaction zone. It is clear that the mechanisms behave in a similar manner regarding the prediction of a single reaction zone structure but differ in determining a double zone structure. They differ in predicting the position of the rich premixed reaction zone that is synergistically attached to the nonpremixed zone.

![Figure 3.20 Comparison of the experimental data and the computational results in the physical space for the temperature profiles for the configuration $\phi_{\text{lean}} = 0.0$ and $\phi_{\text{rich}} = 2.5$ for the Li & Williams Flame.](image-url)
Figure 3.21 Comparison of the experimental data and the computational results in the physical space for the reactant profiles (CH$_4$, O$_2$, N$_2$) for the configuration $\phi_{\text{lean}} = 0.0$ and $\phi_{\text{rich}} = 2.5$.

Figure 3.22 Comparison of the experimental data and the computational results for the products profiles (H$_2$, CO, CO$_2$) in the physical space for the configuration $\phi_{\text{lean}} = 0.0$ and $\phi_{\text{rich}} = 2.5$. 
Figures 3.21 and 3.22 show the respective reactants and products profiles for the case $\phi_{\text{rich}} = 2.5$. The reactant profiles obtained from the simulated results show good agreement with the experimental data. The consumption of oxygen starts at a faster rate on the lean side. These differences are not prominent. The product profiles show some differences in the prediction of the maximum H$_2$ concentrations. The experimentalist had difficulties in the gas-chromatographic analysis of H$_2$ [106]. The CO and CO$_2$ profiles show good agreement with the experimental profiles, although the maximum of experimental CO$_2$ profiles lies above the computed profile. It is interesting to note small differences in the computational results using different versions of the GRI-Mech reaction mechanism. The computations using GRI-Mech 3.0 show better agreement for the products with the experimental data as compared with the results obtained by GRI-Mech 2.11 reaction mechanism.

Figure 3.23 shows the C$_2$H$_6$ concentration profiles for the case $\phi_{\text{rich}} = 2.5$. In the following figure, additionally to the profiles obtained with GRI 2.11 and GRI 3.0 reaction mechanism, the C$_2$H$_6$ concentration profiles are also shown for the CECR and CEC reaction mechanism. It can be observed that all the tested mechanisms are unable to predict the position of the peak concentration. Moreover, the position of the peak concentration is more or less same, as obtained by several mechanisms. The peak C$_2$H$_6$ concentration is overpredicted by the GRI 2.11 and GRI 3.0 reaction mechanism. The profiles of the concentration of C$_2$H$_6$ is similar.
for both the version of GRI mechanism. The reactions for the C₂ - chemistry is primarily same for all the mechanisms. CEC mechanism underpredicts the peak concentration. CECR mechanism predictions of the peak concentration is better as compared to the other mechanisms predictions. Our predictions of the C₂H₆ concentration profile using CECR mechanism is similar to that obtained by Williams et al. [106].

3.4.4 Comparison of the experimental data and the computational results - NO emissions

This section compares the NO concentration measurements of Ref. 106 with predictions.

![Graph comparing experimental data and computational results for NO emissions](image)

**Figure 3.24** Comparison of the experimental data and the computational results in the physical space for the NO mole fraction profiles for the configuration ϕ_{lean} = 0.0 and ϕ_{rich} = 1.5 for the Li & Williams Flame.

Figures 3.24 compares the NO concentrations predicted by the four mechanisms with the measurements for the ϕ_{rich} = 1.5 flame [106]. Two important observations can be made. First, the NO peak that is computed using the GRI 2.11 mechanism is in better agreement with the experimental data than the GRI 3.0, CEC, and CECR mechanisms. Some differences are observed for the NO mole fractions at the edges of the flame due to the reburn NO chemistry. The NO reburn chemistry occurs in the fuel rich region, where the CH_i (i ≥1) radicals that are formed react with the NO to form HCN, which then can be reduced to N₂ through the reaction HCN + NO → N₂ + HCO. The NO is produced in the nonpremixed zone and trans-
Laminar Partially Premixed Flames

ported to the rich zone through convection and diffusion. The reburn process helps reduce the overall NO\textsubscript{x} emissions. The authors of different reaction mechanisms are improving this NO chemistry in the rich reaction zone [17], and this is one of the direction for the improvement of the reaction mechanism considered by the research groups, who are involved in the further development of these mechanisms. Second, the NO concentration peak predicted by the GRI 3.0 mechanism is higher that the result using the GRI 2.11 mechanism. The CECR mechanism underpredicts NO. The NO peak concentration predicted by the CEC mechanism is higher compared to the other mechanisms. The difference between the peak NO predicted by the CEC and the GRI 3.0 mechanisms is =20 ppm.

While most mechanisms predict similar temperature profiles, they predict very different NO production. To locate the source of this behavior, the NO production/consumption pathways for the various mechanisms are studied in detail. It has been found that the CEC and the GRI 3.0 mechanisms include detailed NO consumption pathways, while the CECR mechanism contains a simplified one. These NO consumption reactions are important in the rich premixed reaction zone that is attached to the nonpremixed reaction zone. This may be the reason for the better prediction of NO profiles by the GRI 3.0 and the CEC mechanism in the rich premixed reaction region.

![Figure 3.25](image.png)

**Figure 3.25** Comparison of the experimental data and the computational results in the physical space for the NO mole fraction profiles for the configuration $\phi_{\text{lean}}=0.0$ and $\phi_{\text{rich}}=2.5$ for the Li & Williams Flame.

Figure 3.25 shows NO concentration profiles calculated by the reaction mechanisms and compared with the experimental data of Williams and Li [106] for the $\phi_{\text{rich}}=2.5$ case. In this case none of the mechanisms predict experimental NO
concentration profiles correctly. The GRI 3.0 and CEC mechanisms overestimate the peak NO concentration, while the GRI 2.11 and CECR mechanisms underestimate it. The CEC reaction mechanism is unable to accurately simulate the NO profiles for the Li and Williams measurements [106]. The NO pathway analysis for that configuration shows that in the CEC mechanism, the following reactions

- HNO + OH \to H_2O + NO
- NH + OH \to NO + H_2
- N + OH \to NO + H

are the main contributors to NO production, while in the GRI 3.0 mechanism, the following reactions

- N + OH \to NO + H
- HNO + H \to NO + H_2

have the largest contribution. The mechanisms show differences in predicting the NO concentration in partially premixed flames that have both the single as well as the double reaction zone structure.

![Figure 3.26 Total production rate profiles for CH in the physical space for the configuration $\phi_{\text{lean}} = 0.0$ and $\phi_{\text{rich}} = 1.5$ for the Li & Will- iams Flame using GRI 3.0, CECR and CEC reaction mechanism.](image)

The NO\textsubscript{x} emission index was calculated for the Li and Williams flames. The GRI 3.0 reaction mechanism predicts a higher emission index for the $\phi_{\text{rich}} = 2.5$ flame as compared to the $\phi_{\text{rich}} = 1.5$ case, although the opposite trend is predicted by
using the GRI 2.11 mechanism. The GRI 3.0 mechanism includes more detailed NO formation chemistry as compared to the GRI 2.11 reaction mechanism. The CH kinetics, which are important for prompt NO formation, have been altered in GRI 3.0 mechanism as compared to its preceding mechanism. The NO emission using the GRI 3.0 reaction mechanism for the various equivalence ratios qualitatively reproduces the measured trend [31]. However, the total NO production computed by the GRI 3.0 mechanism is higher than that simulated by using the other mechanisms. This is attributed to the higher production of the CH radicals that play an important role in the NO pathway in the rich premixed reaction zone.

Figure 3.26 presents the CH production rate profiles for the $\phi_{\text{rich}} = 1.5$ flame, using the GRI 3.0, CECR and CEC reaction mechanisms. The GRI 3.0 reaction mechanism predicts a factor of ten higher CH production rate compared with that obtained using the CECR mechanism. The peak CH production rate obtained by the CEC mechanism lies between that calculated by the other two mechanisms. The positions of the CH production/consumption peaks for the CECR and CEC reaction mechanisms are similar. There are very few experimental measurements of CH available. Blauwens et al. [14] measured CH by means of molecular-beam sampling and mass spectrometric detection in methane/air flames, but here is a need for experimental data in rich partially premixed flames. Ravikrishna et al. [83] have done Laser Induced Fluorescence (LIF) measurements and modelling of Nitric Oxide in methane- and ethane/air counterflow diffusion flames. They found that the GRI 2.11 mechanism underpredict the peak NO in all flames. Their result indicates that a refinement of both the prompt-NO and CH kinetics is required for better predictions. The CH-kinetics is still not properly refined in the latest version of the GRI reaction mechanism, i.e. GRI 3.0.

The lower prediction of the NO emission index in the $\phi_{\text{rich}} = 1.5$ flame as compared with the $\phi_{\text{rich}} = 2.5$ case is due to the double reaction zone structure for this configuration. There is not sufficient oxygen available in the rich reaction zone to react with nitrogen to produce NO. Moreover the CH radicals in the rich mixture react with NO leading to the formation of HCN and eventually of $N_2$.

### 3.5 Sensitivity analysis of the reaction mechanisms

Sensitivity analysis of the reaction mechanisms were carried out to reveal the origins of the differing predictions by the various reaction mechanisms regarding the double reaction zone structure. The CECR reaction mechanism is able to predict a broader flame width and yield better predictions for the double reaction zone structure as compared with the predictions of the other tested mechanisms. Since the predictions of the flame structure for the GRI 3.0, GRI 2.11 and CEC reaction mechanisms are similar, we have performed sensitivity and rate of production analyses for the GRI 3.0 reaction mechanism and the CECR reaction mechanism with respect to the stable species at various temperatures.
An analysis of the reactions on the rich premixed side is performed with respect to their overall sensitivity to the net production and consumption of CH$_4$, H$_2$, CO, and CO$_2$ for the $\phi_{\text{rich}} = 1.5$ flame. The corresponding lean-side conditions are maintained at $\phi_{\text{lean}} = 0$ (i.e., pure air) for the counter flow burner [106]. Table 3.2 and 3.3 present results at a temperature of 700 K. (This temperature is selected, since methane and air consumption are initiated at about 700 K [29]). Only the reaction numbers are mentioned in the discussion below, and details of the reactions are available in Tables 1a and 1b. The most significant reactions are shown in the tables. The most significant reactions at this temperature using both the reaction mechanisms are similar and we are unable to find any suspicious reaction that may cause the shift in the position of the rich premixed reaction zone attached to the nonpremixed reaction zone.

<table>
<thead>
<tr>
<th>No.</th>
<th>Overall Sensitivity</th>
<th>#</th>
<th>Reaction</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>5.08479E-01</td>
<td>75</td>
<td>H+O$_2$=&gt;O+OH</td>
</tr>
<tr>
<td>A2</td>
<td>2.98751E-01</td>
<td>103</td>
<td>H+CH$_3$(+M)=&gt;CH$_4$(+M)</td>
</tr>
<tr>
<td>A3</td>
<td>2.34132E-01</td>
<td>197</td>
<td>OH+CO=&gt;H+CO$_2$</td>
</tr>
<tr>
<td>A4</td>
<td>1.93761E-01</td>
<td>167</td>
<td>OH+H$_2$=&gt;H+H$_2$O</td>
</tr>
<tr>
<td>A5</td>
<td>1.18915E-01</td>
<td>168</td>
<td>H+H$_2$O=&gt;OH+H$_2$</td>
</tr>
</tbody>
</table>

Table 3.2: Overall sensitivity coefficients for significant reactions for a flame established at $\phi_{\text{rich}} = 1.5$ and $\phi_{\text{lean}} = 0.0$ using GRI 3.0 reaction mechanism.

<table>
<thead>
<tr>
<th>No.</th>
<th>Overall Sensitivity</th>
<th>#</th>
<th>Reaction</th>
</tr>
</thead>
<tbody>
<tr>
<td>B1</td>
<td>1.03129E+00</td>
<td>1</td>
<td>H+O$_2$=&gt;OH+O</td>
</tr>
<tr>
<td>B2</td>
<td>4.31983E-01</td>
<td>83</td>
<td>CH$_3$+H(+M)=&gt;CH$_4$(+M)</td>
</tr>
<tr>
<td>B3</td>
<td>2.42832E-01</td>
<td>34</td>
<td>CO+OH=&gt;CO$_2$+H</td>
</tr>
<tr>
<td>B4</td>
<td>2.05936E-01</td>
<td>5</td>
<td>H$_2$+OH=&gt;H$_2$O+H</td>
</tr>
<tr>
<td>B5</td>
<td>1.10182E-01</td>
<td>6</td>
<td>H$_2$O+H=&gt;H$_2$+OH</td>
</tr>
</tbody>
</table>

Table 3.3: Overall sensitivity coefficients for significant reactions for a flame established at $\phi_{\text{rich}} = 1.5$ and $\phi_{\text{lean}} = 0.0$ using CECR reaction mechanism.
We next performed a sensitivity analysis with respect to H atoms, which diffuses faster than any other radical. It plays an important role in the initiation of the methane/air reactions. Tables 3.4 and 3.5 present results of such an analysis for the two mechanisms at 700 K for the $\phi_{\text{rich}} = 1.5$ flame. The reactions are shown in the order of decreasing significance. The reactions at the lower positions are not significant compared to the shown reactions. The GRI 3.0 C9 reaction which involves C$_2$ species is not significant in the CECR reaction mechanism and, conversely, reaction D8 in the CECR reaction mechanism is not important in the GRI 3.0 mechanism.

<table>
<thead>
<tr>
<th>No.</th>
<th>Overall Sensitivity</th>
<th>#</th>
<th>Reaction</th>
</tr>
</thead>
<tbody>
<tr>
<td>C1</td>
<td>7.95681E+00</td>
<td>75</td>
<td>H+O$_2$ =&gt; O+OH</td>
</tr>
<tr>
<td>C2</td>
<td>-4.71546E+00</td>
<td>103</td>
<td>H+CH$_3$(+M) =&gt; CH$_4$(+M)</td>
</tr>
<tr>
<td>C3</td>
<td>-2.56885E+00</td>
<td>105</td>
<td>H+CH$_4$ =&gt; CH$_3$+H$_2$</td>
</tr>
<tr>
<td>C4</td>
<td>1.77450E+00</td>
<td>237</td>
<td>HO$_2$+CH$_3$ =&gt; OH+CH$_3$O</td>
</tr>
<tr>
<td>C5</td>
<td>-1.49981E+00</td>
<td>314</td>
<td>2CH$_3$(+M) =&gt; C$_2$H$_6$(+M)</td>
</tr>
<tr>
<td>C6</td>
<td>1.32646E+00</td>
<td>106</td>
<td>CH$_3$+H$_2$ =&gt; H+CH$_4$</td>
</tr>
<tr>
<td>C7</td>
<td>-1.26269E+00</td>
<td>195</td>
<td>OH+CH$_4$ =&gt; CH$_3$+H$_2$O</td>
</tr>
<tr>
<td>C8</td>
<td>1.20744E+00</td>
<td>316</td>
<td>2CH$_3$ =&gt; H+C$_2$H$_5$</td>
</tr>
<tr>
<td>C9</td>
<td>-1.19683E+00</td>
<td>317</td>
<td>H+C$_2$H$_5$ =&gt; 2CH$_3$</td>
</tr>
<tr>
<td>C10</td>
<td>1.19456E+00</td>
<td>197</td>
<td>OH+CO =&gt; H+CO$_2$</td>
</tr>
</tbody>
</table>

Table 3.4: Sensitivity analysis for significant reactions with respect to the H atoms using GRI 3.0 reaction mechanism.
The rich side equivalence ratio ($\phi_{\text{rich}} = 1.5$) lies within the flammability limits for methane/air combustion. We simulate the laminar flame speed of this mixture to determine those reactions that cause different predictions of the position of the rich premixed reaction zone. The laminar flame speed is computed using the Sandia PREMIX code [37]. The calculated laminar flame speed for a rich mixture of $\phi_{\text{rich}} = 1.5$ at 298K and 1 atm pressure using the GRI 3.0 reaction mechanism is 7.2 cm s$^{-1}$ and using the CECR mechanism is 10.04 cm/sec. Experimental data reveal that the laminar flame speed for the mixture should lie between 10 cm s$^{-1}$ and 12 cm s$^{-1}$ [106, 102, 110]. The GRI 3.0 reaction mechanism underpredicts the laminar flame speed, while the CECR mechanism predicts the experimental value with reasonable accuracy. This difference in the laminar flame speed prediction is another instance of the different predictions due to these mechanisms. As mentioned earlier, the CECR reaction mechanism also provides a better prediction of the double reaction zone structure than the other tested mechanisms.
The prediction of the rich premixed reaction zone structure by the CEC mechanism is similar to that by the both versions of the GRI mechanism.

A sensitivity analysis is also performed with respect to the laminar burning speed. Figures 3.27 and 3.28 presented the normalized sensitivity coefficients obtained by using the KINALC program. The first twenty important reactions are included in the figure. We compared the rate constants for reactions in the GRI 3.0 mechanism that are either not significant, or their order of sensitivity is shifted, with reactions in the CECR mechanism. By comparing the Arrhenius plots (of ln(k) versus 1/T) of these reactions we found that the following C_2 reactions might be responsible for the shift in the position of the premixed rich reaction zone and its attachment to the nonpremixed reaction zone.

\[ \text{R1} \quad \text{H} + \text{C}_2\text{H}_5 \rightarrow 2\text{CH}_3 \]
\[ \text{R2} \quad \text{C}_2\text{H}_5 (+M) \rightarrow \text{H} + \text{C}_2\text{H}_4 (+M) \]
\[ \text{R3} \quad 2\text{CH}_3 (+M) \rightarrow \text{C}_2\text{H}_6 (+M) \]

Figure 3.27 Normalized Flame velocity sensitivities 'S_jv' with respect to the rate coefficients of the GRI 3.0 reaction mechanism for the \( \phi_{\text{rich}} = 1.5 \) mixture.
The rate coefficients for these reactions in the GRI 3.0 and CECR reaction mechanisms were also compared with the experimental values provided by the NIST chemical kinetic database [69]. Authors of GRI mechanism [17] stress that their mechanism is an optimized one, and any further development of this mechanism is permissible only by a similar optimization. Specially, tuning some of the rate coefficients to reproduce specific experiments is disallowed, since it may diminish the performance of the mechanism in other cases. We have nonetheless attempted to tune the GRI mechanism in order to reproduce the Li and Williams [106] measurements. The purpose of doing so is to demonstrate that changing only a few rate coefficients can significantly improve the results of any simulation that is the result of a complex mechanism. We do not characterize this tuned mechanism as being improved and do not recommend the corresponding rate parameters for use by others.

CECR reaction mechanism gives better prediction as compared with the other tested mechanisms for capturing the correct position of the rich premixed flame attached to the non-premixed flame in the double flame structure, so it is considered as the reference mechanism for the modification of the GRI 3.0 reaction mechanism.

Figures 3.29, 3.30 and 3.31 present plots of the reaction constant $k$ versus the inverse temperature for the above mentioned reactions in the GRI 3.0 and CECR...
mechanisms and for the GRI 3.0 mechanism with our modified rates. NIST fit curves obtained through the experimental data are also drawn in these figures. The slope of ln(k) versus 1/T provides the activation energy for a particular reaction, which determines the speed of the reaction. The curves obtained for reaction R1 for the two mechanisms differ. Reaction R1 in the GRI 3.0 mechanism has a lower activation energy compared with its value in the CECR mechanism. Therefore, there is excess of methyl in the GRI 3.0 mechanism as compared with the CECR mechanism. Methyl is consumed via two important paths. It is either oxidized to CH₂O or follows reaction R3 to form C₂H₆. Since the rich zone has a relatively small availability of oxygen atoms, methyl radicals are largely consumed here through reaction R3. The reactions behave similarly in both mechanisms at temperatures higher than 1300 K. The curve for the modified rate for reaction R1 is parallel to its CECR mechanism counterpart so that the behaviour of R1 in both mechanisms is similar over most of the temperature range. We thereby retard the "excess" production of methyl in the GRI 3.0 mechanism. The slopes of reaction R2 (cf. Figure 3.30) are similar for both the GRI 3.0 and the CECR mechanisms for temperatures greater than 500 K, and the difference in the slopes is small below that temperature. Therefore, the reaction speed should be similar for reaction R2 in both mechanisms. However, we modified the rate constant such that the slope of the R2 reaction should become parallel to the slope of this reaction curve in the CECR mechanism.

![Diagram](image_url)

Figure 3.29 Reaction constant k versus the inverse temperature for the reaction R1 in the CECR, GRI 3.0 and the GRI 3.0 mechanism with our modified rates. The NIST fit curve and the experimental uncertainty limits are also shown.
The curve for reaction R3 (cf. Figure 3.31) for the GRI 3.0 reaction mechanism shows differences from its CECR counterpart only for higher temperatures above 900K. However, the reactions behave similarly in the lower temperature range in both the mechanisms. Reaction R3 is a temperature independent reaction in the CECR mechanism, and the speed of the reaction does not depend on its activation energy, but on the other parameters, e.g. collision factor. However, this reaction has a finite activation energy in the GRI 3.0 mechanism in the higher temperature range above 900 K. The modified reaction curve for R3 is positioned parallel to the corresponding CECR curve by altering the rate constants. The modified reaction rate constants lie within the experimental uncertainty limits after comparing the constants with the NIST experimental database [69]. The upper and the lower uncertainty limits are also mentioned in the figures. The different reaction rate for R1 in the GRI 3.0 and in the CECR reaction mechanism until 1300 K, which influences the production of methyl radicals, may be the determining cause for the shift in the position of the rich premixed reaction zone and its attachment to the nonpremixed reaction zone.

\[ C_2H_5(+M) \rightarrow C_2H_4+H(+M) \]

Figure 3.30 Reaction constant k versus the inverse temperature for the reaction R2 in the CECR, GRI 3.0 and the GRI 3.0 mechanism with our modified rates.
Figure 3.31 Reaction constant $k$ versus the inverse temperature for the reaction R3 in the CECR, GRI 3.0 and the GRI 3.0 mechanism with our modified rates. The NIST fit curve and the experimental uncertainty limits are also shown.

Table 3.6 shows the original and the modified rate constants of the GRI 3.0 mechanism for the above mentioned three reactions. The modified reaction rate constants lie well within the experimental uncertainty limits after comparing the constants with the NIST experimental database [69].

<table>
<thead>
<tr>
<th>Reaction No.</th>
<th>Original GRI 3.0 Arrhenius Constants</th>
<th>GRI 3.0 Modified Arrhenius Constants</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$A$</td>
<td>$n$</td>
</tr>
<tr>
<td>R1</td>
<td>3.191E+18</td>
<td>-1.2146</td>
</tr>
<tr>
<td>R2</td>
<td>3.969E+13</td>
<td>-0.1169</td>
</tr>
<tr>
<td>R3</td>
<td>6.770E+16</td>
<td>-1.18</td>
</tr>
</tbody>
</table>

Table 3.6: The original and the modified rate constants for the mentioned reactions in GRI 3.0 mechanism (The rate constant is defined as: $k = A \times T^n \exp(-E/RT)$)
Figure 3.32 presents a comparison of the Li and Williams measurements for the $\phi_{\text{rich}} = 1.5$ flame with simulations involving the original and modified GRI 3.0 mechanisms. The prediction of the position of the rich premixed reaction zone is slightly better with the modified mechanism than the original GRI 3.0 reaction mechanism. The predictions on the lean side and the peak temperatures are similar using the original and modified rates. The flame simulated with the modified rates is broader than that predicted by the original GRI 3.0 mechanism. This shows that by tuning the rate constants of the above mentioned C2-reactions, it is possible to improve the simulation of the position of the rich premixed reaction zone in a “double reaction zone” structure. As we have mentioned earlier, an accurate prediction of the double reaction zone structure is important for the better prediction of the rates of the NO production/consumption on rich side of partially premixed flames. The modified mechanism is able to predict the better structure but not accurate and more experimental work is required to do accurate prediction of the flame structure.

Figure 3.33 presents a comparison of the simulated temperature profiles with the Li and Williams measurements for the $\phi_{\text{rich}} = 2.5$ flame, which features a single reaction zone structure. The modified GRI 3.0 reaction mechanism is able to predict a single reaction zone structure as predicted by the original GRI 3.0 reaction mechanism. This indicates that the reactions involving C2 species play an important role in the determination of the position of the rich premixed reaction zone and its attachment to the nonpremixed reaction zone, but do not influence the predictions of the lean side flame structure or the maximum peak temperature. Li and
Williams [106] were unable to predict the C2 concentration profiles using their mechanism as mentioned in the earlier section.

The tested reaction mechanisms exhibit different results for the NO concentration (cf. Figure 3.24, 3.25). One cause for the differences may be related to the different predictions of the CH concentration profile (cf. Figure 3.26) by the various mechanisms. The CH radical plays an important role in the prompt mechanism, which is dominant on the rich side. In order to study the NO chemistry in further detail, a sensitivity analysis of the CECR and GRI 3.0 mechanisms was also performed. Table 3.7 and Table 3.8 present the results of the overall sensitivity analysis with respect to NO and NO2 for the mechanisms at 1200K, that corresponds to the prompt mechanism initiation temperature [58], and a 1 atm pressure for the \( \phi_{\text{rich}} = 1.5 \) flame. Miller et al. [58] have investigated the prompt initiation reaction (CH + N2 \( \rightarrow \) HCN + N) in the different temperature range from 1000K till 4000K. They provided the information of the rate constants for the mentioned reaction in the temperature range of 1000K till 2000K. The first ten most significant reactions that influence the production and consumption of the NO and NO2 are included in the tables. The C2 – reactions identified previously in context of the double reaction zone structure also play an important role in the production and consumption of NO and NO2. While these reactions are significant in the GRI 3.0 mechanism, they are not so important in the CECR reaction mechanism. This might be a cause for the underprediction of the NO concentration (cf. Figures 3.24, 3.25) by the CECR mechanism.

![Figure 3.33 Comparison of the experimental data and the computational results in the physical space for the temperature profiles for the configuration \( \phi_{\text{lean}} = 0.0 \) and \( \phi_{\text{rich}} = 2.5 \) for the Li & Williams Flame showing the prediction of the modified reaction mechanism.](image)
### Table 3.7: Sensitivity analysis for significant reactions with respect to the NO/NO₂ species using GRI 3.0 reaction mechanism.

<table>
<thead>
<tr>
<th>No.</th>
<th>NO/NO₂ sens.</th>
<th>#</th>
<th>Reaction</th>
</tr>
</thead>
<tbody>
<tr>
<td>E1</td>
<td>5.01854E+01</td>
<td>75</td>
<td>H+O₂ =&gt; O+OH</td>
</tr>
<tr>
<td>E2</td>
<td>1.14996E+01</td>
<td>103</td>
<td>H+CH₃(+M) =&gt; CH₄(+M)</td>
</tr>
<tr>
<td>E3</td>
<td>6.14430E+00</td>
<td>105</td>
<td>H+CH₄ =&gt; CH₃+H₂</td>
</tr>
<tr>
<td>E4</td>
<td>1.66526E+00</td>
<td>237</td>
<td>H₂O₂+CH₃ =&gt; OH+CH₃O</td>
</tr>
<tr>
<td>E5</td>
<td>1.60933E+00</td>
<td>106</td>
<td>CH₃+H₂ =&gt; H+CH₄</td>
</tr>
<tr>
<td>E6</td>
<td>1.52406E+00</td>
<td>478</td>
<td>CH+N₂ =&gt; HCN+N</td>
</tr>
<tr>
<td>E7</td>
<td>1.35685E+00</td>
<td>197</td>
<td>OH+CO =&gt; H+CO₂</td>
</tr>
<tr>
<td>E8</td>
<td>1.06482E+00</td>
<td>314</td>
<td>2CH₃(+M) =&gt; C₂H₆(+M)</td>
</tr>
<tr>
<td>E9</td>
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<td>148</td>
<td>C₂H₅(+M) =&gt; H+C₂H₄(+M)</td>
</tr>
<tr>
<td>E10</td>
<td>8.97431E-01</td>
<td>317</td>
<td>H+C₂H₅ =&gt; 2CH₃</td>
</tr>
</tbody>
</table>

### Table 3.8: Sensitivity analysis for significant reactions with respect to the NO/NO₂ species using GRI 3.0 reaction mechanism.

<table>
<thead>
<tr>
<th>No.</th>
<th>NO/NO₂ sens.</th>
<th>#</th>
<th>Reaction</th>
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<tr>
<td>F1</td>
<td>1.69099E+02</td>
<td>1</td>
<td>H+O₂ =&gt; OH+O</td>
</tr>
<tr>
<td>F2</td>
<td>3.68230E+01</td>
<td>83</td>
<td>CH₃+H(+M) =&gt; CH₄(+M)</td>
</tr>
<tr>
<td>F3</td>
<td>2.46288E+01</td>
<td>57</td>
<td>CH₄+H =&gt; H₂+CH₃</td>
</tr>
<tr>
<td>F4</td>
<td>6.15735E+00</td>
<td>58</td>
<td>H₂+CH₃ =&gt; CH₄+H</td>
</tr>
<tr>
<td>F5</td>
<td>2.42075E+00</td>
<td>107</td>
<td>CH₂+O₂ =&gt; CO+OH+H</td>
</tr>
<tr>
<td>F6</td>
<td>2.38222E+00</td>
<td>34</td>
<td>CO+OH =&gt; CO₂+H</td>
</tr>
<tr>
<td>F7</td>
<td>2.08779E+00</td>
<td>81</td>
<td>2CH₃ =&gt; C₂H₅+H</td>
</tr>
<tr>
<td>F8</td>
<td>1.97454E+00</td>
<td>59</td>
<td>CH₄+OH =&gt; H₂O+CH₃</td>
</tr>
</tbody>
</table>
Table 3.8: Sensitivity analysis for significant reactions with respect to the NO/NO$_2$ species using GRI 3.0 reaction mechanism.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Rate Coefficient</th>
<th>Species</th>
<th>Reaction</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH$_3$+HO$_2$=&gt;CH$_3$O+OH</td>
<td>1.94337E+00</td>
<td>73</td>
<td>F9</td>
</tr>
<tr>
<td>2CH$_3$(+M)=&gt;C$_2$H$_6$(+M)</td>
<td>1.78429E+00</td>
<td>85</td>
<td>F10</td>
</tr>
</tbody>
</table>

Figures 3.34 and 3.35 present comparisons of the NO concentration profiles simulated by the original GRI 3.0 reaction mechanism and the modified rate GRI 3.0 mechanism with the experimental data of Li and Williams [106]. The predictions of the peak NO concentrations by the modified GRI 3.0 reaction mechanism are in better agreement than the predictions by the original GRI 3.0 reaction mechanism in both flames. Also, the slope of the NO profile in $\phi_{\text{rich}} = 1.5$ case (cf. Figure 3.34) is captured by the modified mechanism better than that by the original GRI 3.0 mechanism on the rich side. The behaviour of both mechanisms for NO production/consumption on the lean side is similar as depicted by the NO concentration slopes (cf. Figure 3.34,3.35). This is to be expected, since we have only altered the abovementioned C$_2$ – reactions, which impact the NO chemistry on...
the rich side. The predictions of the modified GRI 3.0 mechanism are better compared to the other tested mechanisms in this regard (cf. Figures 3.34, 3.35). The C2-reactions identified previously play an important role in obtaining better predictions of NO emissions in these rich partially premixed flames.

![Graph showing NO concentration profiles](image)

**Figure 3.35** Comparison of the experimental data and the computational results in the physical space for the NO mole fraction profiles for the configuration $\phi_{\text{lean}} = 0.0$ and $\phi_{\text{rich}} = 2.5$ for the Li & Williams Flame showing the predictions of the modified reaction mechanism.

### 3.6 Conclusions from this section

Atmospheric laminar partially premixed counterflow flames are investigated here. The double structure of the partially premixed flame leads to lower NO emissions, provided that a desirable strain rate is available. The effects of strain rate on NO production rate are investigated. At lower strain rates the partially premixed flame stabilizes along the non-premixed flame and this leads to NO production primarily in the non-premixed reaction zone. In the rich premixed flame, there is relatively little oxygen available for NO production, provided the strain rate is favourable. On the other hand, at higher strain rates partially premixed flames have a lower residence time for stabilization, due to the higher convective trans-
Laminar Partially Premixed Flames

port of the unburned mixture as compared to the diffusive transport of heat from the nonpremixed flame. Consequently, a merged premixed-nonpremixed flame exists. This leads to higher NO production due to both the prompt and thermal NO mechanisms. Comparisons of the present results with experimental data available from the literature are presented. The simulated results obtained by using various reaction mechanisms exhibit similar behavior with respect to the stable species and temperature profiles regarding the prediction of a single flame structure. On the other hand, differences are observed for the NO chemistry. Another problem is encountered regarding the rich side chemistry after comparing the simulations with experimental data. The various mechanisms show differences in their predictions of the position of the rich premixed reaction zone and its attachment to the nonpremixed reaction zone that leads to a double reaction zone structure. An analysis of the reactions is performed and reactions that might cause this discrepancy are identified. The reaction rates are modified for the GRI 3.0 mechanism to obtain a better prediction of the thickness of the reaction zones and the double reaction zone structure. Predictions of NO concentration are also better with the modified mechanism. However, in order to justify this modification, representative measurements of C₂ reactions and also CH concentration are required.
4 Turbulent Partially Premixed Flames

This section gives an overview about the research that has been carried out to understand the structure of the turbulent partially premixed flames. The effect of the partial premixing on the NO\textsubscript{x} emissions are discussed for these flames. The computational models suggested for the description of such flames are already mentioned in the earlier section (section 2.4.1). Here the transient flamelet model will be described, which was developed by Ferreira et al. [26]. It has been explained earlier that the diffusion based combustion model can be developed using the transported scalar, named mixture fraction, in the flow field. The diffusion combustion is mixing controlled and the mixture fraction scalar determines the extent of the mixing. In a similar manner the premix combustion is reaction controlled and the model can be developed with the help of the scalar, called reaction progress variable. The progress of the reaction is described by the reaction progress variable. The concept of using the existing model for predicting the turbulent partially premixed flame is explained. The limitations of the existing flamelet libraries to describe the partially premixed flame are identified. The transient laminar flamelet libraries are modified and generated in the present work to account for the partially premixed phenomenon. Moreover, a certain type of transformation into the mixture fraction space of the diffusion flame is done in the transient flamelet code to adapt the pure diffusion flamelet to the partially premixed case. This is explained in the latter section (Section 4.3.1). A partially premixed turbulent flame using the existing model with new transient flamelet libraries is computed. The results and the shortcomings of the existing model will be discussed in the next chapter.

4.1 Literature review

Bilger et al. [54] investigated turbulent non-premixed flames of methane near extinction. During that period the measurements of the fluctuations are not available, so they want to distinguish whether the measured change in the flame structure as it approaches extinction is due to unmixness or unreactedness or both. They observed that in the fixed axial location, the unmixedness profile remains almost unchanged with the increasing mean jet velocity. Hence, they concluded that change in the flame structure is entirely due to unreactedness. Further they also found that the concept of the existence of partially premixed laminar diffusion flamelets can not be completely excluded from such type of flames.

Peters et al. [74] have also suggested the structure of partially premixed diffusion flamelets in a turbulent flow field. They observed that such type of structures can exist if the variation of the mixture fraction is quite large.

Mansour et al. [51] carried out simultaneously Raman/Rayleigh measurements in turbulent partially premixed flames of nitrogen diluted methane near extinction. They studied the effects of extinction on the flame structure. They found that the reaction progress decreases at stoichiometric mixture fraction near the extinction.
However, the reactedness occurs around the mean stoichiometric of the flame. They observed the chemical kinetic effects in these flames on approaching the extinction, thereby reducing the residence time. Due to the chemical effects, the product mass fraction and the peak temperature reduces and the reactant mass fraction increases.

Mastorakos et al. [55] have done experiments on turbulent counterflow diffusion flames with partial premixing. They stabilized the natural gas flames between two opposing jets. They observed that the extinction strain rate increased from 350 sec\(^{-1}\) for pure fuel to 600 sec\(^{-1}\) for the partially premixed mixture of \(\phi = 2.4\). These values were found to be similar to those measured by them at extinction of laminar counterflow strained flames. This suggests that turbulent non-premixed flames extinguish at a critical strain rate equal to the critical strain rate for extinction of laminar counterflow flames. They have also shown that partial premixing makes the turbulent flame more strain resistant. Similar results are also obtained by Puri et al. [91] for the laminar counterflow flames.

Turns et al. [100] studied experimentally the effects of the partial premixing on the turbulent jet flames. They studied turbulent partially premixed methane-, propane- and ethylene/air flames. For burning propane and methane with a decreasing value of \(\phi_{\text{fuel}}\) (fuel equivalence ratio), the EINO\(_x\) (the NO\(_x\) emission index) first increases and for smaller values of \(\phi_{\text{fuel}}\) decreases again. This behaviour is believed to be a consequence of the competing effects of decreasing radiative heat loss and declining residence times as more air is added to the fuel owing the reduced soot formation.

Hustad et al. [86] studied the unconfined propane-air turbulent partially premixed coflow flames. This experimental investigation is carried out to validate various correlations for liftoff flames from previous studies focused on diffusion flames. They proposed a new correlation for the liftoff of the partially premixed propane-air turbulent flames. They also analysed the existing correlation for NO\(_x\) emissions from turbulent free jet diffusion flames based on flame volume and fuel mass flow. They modified this correlation by introducing the fuel mass fraction to the existing correlation for the predictions of the NO\(_x\) emissions. Their findings show that the emissions of the NO\(_x\) increases by increasing the air concentration in the partial premixed jet stream. Their conclusion necessitated the need to study the structure of the partially premixed flames for the predictions of the NO\(_x\) emissions.

Peters et al. [52] studied experimentally the partially premixed turbulent jet flames stabilized by hot combustion products. They used the equivalence ratio of 2 for the structure analysis of the partially premixed flames. They found that the mixture fraction and the progress variable based on the mass fraction of H\(_2\)O is necessary for the detailed description of structure of the partially premixed flames. A partially premixed piloted jet flame [5] using the transient flamelet model is computed in the present work. It is not possible to describe this flame using the standard diffusion flamelet model involving only mixture fraction variable and scalar dissipation rate.

Gore et al. [49] carried out experiments in order to study the pollutant emission characteristics of partially premixed turbulent jet flames. Emission indices for NO,
NO\(_X\), CO and HC were measured over a broad range of \(\phi_B\) from infinity to unity, which includes both limiting cases of diffusion and fully premixed flames. The EINO\(_X\) behaviour was divided into three sections for both confined and unconfined flames: (1) a constant EINO\(_X\) for a fixed fuel rate in the range of 5<\(\phi_B<\infty\), (2) a decrease of the EINO\(_X\) of about 25% for the range of 1.5<\(\phi_B<5\) reaching a minimum at \(\phi_B=1.5\), (3) a steep increase for the EINO\(_X\) at 1.0<\(\phi_B<1.5\) peaking at \(\phi_B=1\), the fully premixed flame. Measurements of the visible flame length led to the conclusion that the residence time could not explain solely the EINO\(_X\) trend. A similar result was obtained for the radiative heat loss fraction. Although measurements of temperature profiles give possible explanations for the ranges of 2<\(\phi_B<\infty\) and 1.0<\(\phi_B<1.5\), the decrease in EINO\(_X\) for reducing \(\phi_B\) in the range 1.5<\(\phi_B<2.0\) cannot be explained by the above mechanisms. They attributed this behaviour of the emission index for changes to the contributions of either the prompt or the reburn mechanism, which corresponds to our own findings [16].

There has not been much work done in the modeling of the turbulent partially premixed flames. As mentioned in the chapter 2, it has been shown that the structure of the partially premixed flames are similar to the diffusion flames. Moreover, the models developed to predict the turbulent premixed flames and diffusion flames, can be combined to describe the structure of the turbulent partially premixed flames.

In the following section the concept of using the transient flamelet model for the prediction of the turbulent partially premixed flames is explained. This model is developed by Ferreira et al. [24] to predict the lifted turbulent diffusion flames.

4.2 Modeling approach for turbulent partially premixed flames

The concept of the flamelet approach has already been discussed in the earlier section. The existing transient flamelet model contains the following four variables for describing the lifted turbulent diffusion flames, namely

- Mixture fraction
- Scalar dissipation rate
- Flamelet evolution time
- Reaction progress variable

The mixture fraction is used to describe the mixing phenomenon, which is determinant for describing the diffusion flames. The scalar dissipation rate accounts for the impact of the turbulence on the combustion. The flamelet evolution time accounts for the development of the flamelets in the mixture fraction space. The reaction progress variable is used to describe the reignition phenomenon in diffusion flames after a certain time has been elapsed.

The partially premixed flame contains both the non-premixed and the premixed modes of combustion. So the above mentioned scalars are required for describ-
ing the partially premixed flames [60] [85]. In the present work the capability of the existing flamelet model is adapted to the partially premixed case and also tested.

### 4.2.1 Concept

It has been explained earlier that the partially premixed flames can be described using the above-mentioned four scalars. However, the question arises, what are the limitations of the existing flamelet model for describing the partially premixed turbulent flames. It has been observed [11] that in most of the industrial flames the structure of the turbulent partially premixed flames is similar to the turbulent diffusion flames. These similar structures are observed only if the equivalence ratio of the fuel stream is above a critical value or the flow is highly turbulent. This can be explained in the next paragraph with respect to the flame structure of the counterflow laminar partially premixed flames.

The earlier figures (cf. Figures 3.2 and 3.3 from the section on laminar flames) show the flame structure of the counterflow laminar partially premixed flames along the axis for different equivalence ratio. These figures are again shown here for the convenience of the reader (Figures 4.1 and 4.2).

It is observed that the structure of the flame for the case \( \phi_{\text{rich}} = 2.7 \) and \( \phi_{\text{lean}} = 0.0 \) is similar to the diffusion flame and no separate double flame structure is visible. Moreover at high strain rate (high velocity) the flame structure for the case with \( \phi_{\text{rich}} = 1.7 \) and \( \phi_{\text{lean}} = 0.0 \) changes from double structure to single flame structure similar to the non-premixed flame structure (cf. Figure 4.3). Blevins et al. [15] have also observed double flame structure of the partially premixed flames at lower strain rate. These structure merges at higher strain rate to give rise to single flame structure similar to non-premixed flames.

**Figure 4.1** Species mole fraction and temperature profiles in the physical space for \( \phi_a = 0.0 \) and \( \phi_b = 1.7 \)
Figure 4.2  Species mole fraction and temperature profiles in the physical space for $\phi_a=0.0$ and $\phi_b=2.7$

Figure 4.3  Effect of the strain rate on the flame structure of laminar partially premixed counterflow flame
Turbulent Partially Premixed Flames

Figure 4.4 Temperature profiles in the physical space for $\phi_{\text{rich}} = 2.8$, $\phi_{\text{rich}} = \infty$, and $\phi_{\text{lean}} = 0.0$ for both the cases.

Figure 4.4 shows the temperature profile for the counterflow laminar diffusion flame and the partially premixed ($\phi_{\text{rich}} = 2.8$, $\phi_{\text{lean}} = 0.0$) laminar flame in the physical space. The distance between the fuel and the air nozzle is 1 cm. The computations are carried out using the GRI 3.0 reaction mechanism. It can be observed that the flame structure for the partially premixed flame is similar to the diffusion flame structure in the physical space. However, some shift is observed in the peak flame temperature.

Figure 4.5 shows the species profiles (H$_2$ and H$_2$O mole fraction) for the diffusion flame and the partially premixed laminar flame. Again, the species concentration profiles are observed to be similar for both the flames in the physical space. Some differences are observed with respect to the position of the peak species concentration in the physical space. However, these differences should not be so significant in the turbulent flames.

The difference in the peak temperature and the peak species concentration is due to the effect of the partial premixing on the reactedness.
In the transient flamelet model, the flame structure in the flamelet library is based on the mixture fraction space. The unsteady governing equation of species and the energy is solved in the mixture fraction space to generate the flamelet libraries [26]. These equations are known as Peter's Equations. The transformation of the species and the energy equations from the physical space to the mixture fraction space was first given by N. Peters. The species and the energy equations in the mixture fraction space are written below:

\[ \frac{\partial Y_n}{\partial t} = \chi \frac{1}{2 L e_n} \left( \frac{\partial^2 Y_n}{\partial Z^2} \right) + \frac{\dot{\omega}_n}{\rho} \]  

(4.1) 

\[ c_p \frac{\partial T}{\partial t} = \chi \left( \frac{\partial^2 h}{\partial Z^2} \right) - \sum_{n=1}^{N} h_n \left( \frac{\chi}{2} \frac{\partial Y_n}{\partial Z} + \frac{\dot{\omega}_n}{\rho} \right) \]  

(4.2) 

The species concentration values to be delivered for a particular local condition in the turbulent flow field is taken from the transient flamelet library based on the value of the mixture fraction, scalar dissipation rate, turbulence time scale and the reaction progress variable. So the above shown figures (cf. Fig. 4.4) are redrawn in the mixture fraction space to estimate the error involved in using the transient flamelet library based on the pure diffusion laminar flame for the description of the turbulent partially premixed flame.

Figure 4.6 shows the temperature profile in the mixture fraction space for both the case of diffusion and the partially premixed laminar flame. The mixture fraction is defined using the molecular nitrogen.
We assume that the combustion takes place at the stoichiometric mixture fraction in the laminar flames. The peak temperature in the mixture fraction space at the stoichiometry for the diffusion flame is 1940 K and the peak temperature for the partially premixed flame is 1830K. The error introduced by using the diffusion flamelet library for the description of the peak temperature in the partially premixed flame is 6% for the present configuration of the partially premixed flame. The position of the peak temperature is the same for both the flames. The error in the peak mass fraction of H₂O species is about 10% at the stoichiometry by using these libraries for predicting the partially premixed flames. It has been assumed that the combustion takes place at the condition of stoichiometry. The error in describing the magnitude of the species value increases with the increase in the deviation of the mixture fraction from the stoichiometric mixture fraction on the rich mixture side.

It has to be noted that the local conditions corresponding to the rich mixture in the flow field (for the case with mixture fraction greater than stoichiometric mixture fraction) can not be derived from the transient flamelet library. The transient flamelet library, as mentioned earlier, is based on the diffusion flame. So there is a requirement of doing a transformation of the mixture fraction in the flow field to the mixture fraction space in the transient flamelet library. This is explained later in this chapter.

Figure 4.6 Temperature profiles in the mixture fraction space (based on N₂) for φ_{rich} = 2.8, φ_{rich} = ∞ and φ_{lean} = 0.0 for both the cases.
Figure 4.7 Species (H₂O) profiles in the mixture fraction space (based on N₂) for \( \phi_{\text{rich}} = 2.8, \phi_{\text{rich}} = \infty \) and \( \phi_{\text{lean}} = 0.0 \) for both the cases

It can be concluded from the above observation that the flame structure of the partially premixed flame is similar to the non-premixed flame in the physical space as long as the equivalence ratio of the fuel stream is rich enough to give rise to the single flame structure similar to the nonpremixed flame. Therefore the transient laminar flamelet model developed for predicting the lifted diffusion flames, should be able to predict the diffusion type partially premixed turbulent flames. However, it has to be noted that the magnitude of the error in describing the values of the species increases with a increase in the deviation of the mixture fraction in the rich mixture away from the stoichiometric value of mixture fraction. The transformation of the mixture fraction space for the partially premixed flame into the mixture fraction space for the diffusion flame is required to get the correct values of the species from the transient flamelet library.

In order to predict the accurate value of the species concentration, it is required to use a new transient flamelet library based on the boundary condition available for the turbulent flame. e.g. In the present case a new library is required with the partially premixed boundary conditions.

The flamelet libraries developed for the diffusion flame can be utilized in describing the structure of partially premixed flames having structure similar to the diffusion flame. A test case has been computed using the mentioned transient flamelet model. The results are discussed in the next section. Also the error in describing the magnitude of the species concentrations and the temperature are discussed in the next chapter.
4.2.2 Summary of transient flamelet model for the lifted diffusion flames

The following section describes the transient flamelet model for the prediction of the lifted diffusion flames in detail.

In turbulent reacting flows the combined effects of mixing and turbulence can be described by the mixture fraction and the scalar dissipation rate.

The scalar dissipation rate in the laminar flamelets can be classically defined \[75\] as

\[
\chi = 2D\left(\frac{\partial Z}{\partial \chi}\right)^2
\]  

(4.3)

where the constant of proportionality is the mass diffusion coefficient of the species.

The scalar dissipation rate in the turbulent flame is expressed as

\[
\chi = C_\chi \tilde{\varepsilon} \tilde{\alpha}^2
\]  

(4.4)

where \(\tilde{\varepsilon}\) over the variables represents the Favre averaged terms in the turbulent flow field. The averaging methods in the reacting and the non-reacting flows are discussed elsewhere \[24\].

In the above expression the standard value \(C_\chi = 2.0\) is used. The data of the laminar flamelet computations, which are done prior to the turbulent combustion calculations, is stored in prepared laminar flamelet libraries. These libraries are based on the mixture fraction and the scalar dissipation rate for the generic flamelet model for diffusion flames.

The standard laminar flamelet model does not contain any scalar that describes the progress of partially premixed reactants or the intermediate species after extinction. It is essential to have another scalar in addition to the mixture fraction and the scalar dissipation rate to describe the partially premixed combustion \[2\] \[60\], since the partially premixed combustion contains both the diffusion and the premixed combustion modes.

The standard steady flamelet model has been developed \[24\] and incorporated in the commercial CFD code \[62\]. Ferreira \[24\] has extended the steady laminar flamelet model (SLFM) to predict the extinction and the re-ignition phenomenon in the turbulent diffusion flame. This extended model is known as transient laminar flamelet model (TLFM) and contains additional scalars, namely reaction progress variable and the flamelet evolution time. The reaction progress variable describes the re-ignition of the extinguished intermediate species after certain time has elapsed.

Since the TLFM accounts for the progress of the reactions of the intermediate species in the diffusion flame, so this model should be also applicable for predicting the flame structure of the partially premixed turbulent flames. In the present
work, we are interested in testing the capability of the TLFM in its existing form for the predictions of the partially premixed flame. The capability and the shortcoming of the existing model is discussed in the next section.

The transient flamelet library in its present form accounts for only partially premixed stages that describes the re-ignition phenomenon in the turbulent diffusion flame. It will be explained later, that the libraries available are not sufficient to describe the flame structure of turbulent partially premixed flames. So a new set of flamelet libraries for the transient laminar flamelet model have been generated to account for the progress of the partially premixed reactants and thus describing the partially premixed turbulent flame.

In the next paragraphs the governing equations of the transient flamelet model are summarized.

The mixing model based on the approach known as Interaction by Exchange with the Mean (IEM approach) is proposed by Ferreira et al. [24]. This model accounts for the partially premixed states of the reactants.

A small fluid element of uniform composition defined by the composition vector \((Y_n^*, Z^*)\), where \(Y_n^*\) are the mass fractions of the main stable species and \(Z^*\) is the mixture fraction. There is a continuous mixing between this fluid element and other fluid elements through its interface with the surroundings.

Figure 4.8 shows schematically the distribution of the fuel, the oxidizer and the product. If due to high strain rate the combustion extinguishes, which leads to partially premixed reactants. Corresponding to the flamelet evolution time, these extinguished flamelets will reignite and can develop in the composition space, provided the strain rate is lower and enough time is available for the reactions to take place and leads to complete combustion. The initial mixing line after the certain time has been elapsed is shown in figure 4.8. The initial partially premixed mixture is described along this mixing line. In the present model the initial partially premixed mixture takes only the reactants and the products into account. However, the intermediate products are not taken into account. Some of these intermediate species are not stable and the reaction time is so fast that they can be neglected (C2 species). However, the oxidation reaction of CO is slower than the other methane reactions. So the initial partially premixed mixture should contain also CO species along with the reactants and the stable products like H2O and CO2.
The values of $Y_{F,u}^*$, $Y_{O_2}^*$ and $Y_{F,b}^*$ are related to each other by a global reaction step and is known as reaction progress variable $c^*$. Mathematically it can be expressed as:

$$c^* = \frac{Y_{F,u}(Z^*) - Y_F^*}{Y_{F,b}(Z^*) - Y_{F,b}^*} \quad (4.5)$$

where $Y_{F,u}(Z^*)$ and $Y_{F,b}(Z^*)$ are the fuel mass fractions of the completely unburnt and burnt conditions, respectively. For the burnt condition the Burke-Schumann solution is used, although it is possible to use a steady state flamelet solution for the given scalar dissipation rate. Oxidizer and product are then also defined by the reaction progress variable through the following equations:

$$Y_{O_1}^* = Y_{O_1,u}(Z^*) - c^*(Y_{O_1,u}(Z^*) - Y_{O_1,b}(Z^*)) \quad (4.6)$$

$$Y_p^* = c^*Y_{p,b}(Z^*) \quad (4.7)$$
The composition vector which defines the initial mixing state is fully defined by a single reaction progress variable and the mixture fraction, and may be written as \((c^*, Z^*)\).

It is assumed that the highest probability of ignition will occur for fluid elements with a composition near the stoichiometric mixture. Therefore the value of \(Z\) equal to the stoichiometric value is set and the composition vector for partially premixing and its distribution may be written as \((c^*, Z_{st})\).

The mean species mass fractions for the transient flamelet model are obtained from the following expression

\[
\bar{Y}_n = \int_{0}^{1} \int_{0}^{1} \int_{0}^{1} \int_{0}^{1} Y_n(Z, Z^*, c^*, \chi, t_\beta) P(Z, Z^*, c^*, \chi, t_\beta) dZ. dZ^*. dc^*. d\chi. dt_\beta
\] (4.8)

In the above equation, the joint pdf's are not known. If it is assumed that the variables are statistically independent. Then the above equation can be re-written as follows

\[
\bar{Y}_n = \int_{0}^{1} \int_{0}^{1} \int_{0}^{1} Y_n(Z, Z^*, c, \chi, t_\beta) P(Z) \delta(c - c^*). \delta(\chi - \chi^*). \delta(t_\beta - t_\beta) dZ. dc. d\chi. dt
\]

For the variables \(c, \chi\) and \(t_\beta\) \(\delta\)-functions have been chosen as PDF's. The \(\delta\)-function is selected due to its easy implementation in the code and the time restrictions for the implementation of the complex pdf functions like \(\beta\) for the reaction progress variable and log-normal function for the scalar dissipation rate. \(P(Z)\) is represented by the \(\delta\)-function which is given as.

\[
P(Z) = Z^{\alpha-1}(1-Z)^{\beta-1} \frac{\Gamma(\gamma)}{\Gamma(\alpha) \Gamma(\beta)}
\] (4.10)

where \(\Gamma\) is the Gamma function and \(\alpha, \beta,\) and \(\gamma\) are calculated from

\[
\alpha = \tilde{Z}\gamma \quad \beta = (1-\tilde{Z})\gamma \quad \gamma = \frac{\tilde{Z}(1-\tilde{Z})}{\tilde{Z}^\gamma - 1}
\] (4.11)

In the present model the variation of the scalar dissipation rate is not taken into account. The PDF of the scalar dissipation rate should be a log-normal distribution as suggested in [73] in order to predict the effect of the scalar dissipation rate on the reacting flows in a more realistic manner.

The "\(\tau\)" is assumed to be the residence time of the flamelet before it changes its state in the flow field. This time accounts for the transient behaviour of the flamelets and is therefore called as flamelet evolution time. The mean flamelet evolution time is modelled as the characteristic turbulence time:
The equation 4.9 requires the input of the species concentration from the transient laminar flamelet library. This information is stored as a function of mixture fraction, scalar dissipation rates, reaction progress variables and the time scales in the form of tables.

As mentioned in the transient laminar flamelet model an additional transport equation for the mean reaction progress variable \( \bar{c} \) has to be solved in order to consider the existence of non-reacting but partially premixed states. This reaction progress variable can be regarded as a normalised reactive species. Therefore, its transport equation correspond to those of a reactive species.

\[
\frac{\partial \bar{c}}{\partial t} + \frac{\partial \bar{\rho} \bar{u} \bar{c}}{\partial x_j} = \frac{\partial}{\partial x_j} \left( \bar{\rho} D + \frac{\mu_\lambda}{Sc_j} \frac{\partial \bar{c}}{\partial x_j} \right) + \bar{S}_c \tag{4.13}
\]

The source term, which represents the chemical reactions in the turbulent flow field, has to be modelled. In our case the final value of the reaction progress is defined as \( c_{fl} \) within the flamelet library. Therefore, the source term \( S \) can be defined in a way, that the transported value \( \bar{c} \) is approaching the value \( c_{fl} \), as long the latter is higher than \( \bar{c} \)

\[
\bar{S}_c = C \left( c_{fl} - \bar{c} \right) \tag{4.14}
\]

In the case \( c_{fl} < \bar{c} \), the source term is not active, because the local flow condition is not favourable for combustion and a non zero value of \( \bar{c} \) is only due to transport without chemical reaction. The constant \( C \) in equation 4.14 is a large number in order to suit numerical convergence [72].

### 4.2.3 Transient flamelet library

The new flamelet libraries are generated using detailed chemistry and one dimensional laminar code FLATRA [26], which computes the exact transient state solution of the species concentrations and the temperature. The generated libraries are developed using the nonpremixed configuration and is suitable for describing the test case, where the flame structure is similar to the diffusion flame structure.

### 4.3 Discussion on the transient flamelet library

The old transient flamelet libraries are generated to capture the extinction and reignition phenomenon in turbulent diffusion flames. The range of the reaction progress variable assumed for the prediction of the re-ignition phenomenon is not sufficient enough to predict partially premixed turbulent flames. The range earlier
used is selected for the prediction of a lifted diffusion flame [24]. This range of the reaction progress variable and the scalar dissipation rate is shown in the figure 4.9. The reignition phenomenon can take place only above the curve as shown in the figure below. Moreover the limits of the scalar dissipation rate for capturing the flame structure for the simulation of the tested flame is also marked in the figure shown below.

![Figure 4.9 Range of the reaction progress variable in the old transient flamelet library (Figure shows the C_{min.} values versus inverse of scalar dissipation rates).](image)

Figure 4.9 Range of the reaction progress variable in the old transient flamelet library (Figure shows the $C_{\text{min.}}$ values versus inverse of scalar dissipation rates).

However, this range of the reaction progress variable is not sufficient to account for the burning of the partially premixed reactants, which are coming from the fuel/air nozzles. The range shown above in the figure accounts only for reignition of the partially premixed state after certain level of combustion and extinction has taken place and the mixture is still ignitable provided the strain rate is below the quenching strain rate.

The range of the scalar dissipation rate, flamelet evolution time and the reaction progress variable in the transient flamelet library is shown below in the tabulated form:

<table>
<thead>
<tr>
<th>Scalar</th>
<th>Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\chi$ [1/sec]</td>
<td>0.01, 0.1, 1.0, 5.0, 10.0</td>
</tr>
</tbody>
</table>

Table 4.1: Old range of the transient flamelet library
New transient flamelet libraries are generated for the whole range of the reaction progress variable from 0 till 1 with an interval of 0.1. Also the range of the scalar dissipation rate is selected according to the test case turbulent flame [4]. This flame is very turbulent with Reynolds number of the order of 20000 based on the inlet nozzle diameter. The flame structure of this turbulent flame is similar to the diffusion flame.

The existing model with new transient flamelet libraries is used for the computation of the partially premixed turbulent piloted jet flame [5]. The configuration of the geometry, boundary conditions and the results are discussed in the next chapter.

### 4.3.1 Transformation in the Mixture Fraction Space

As mentioned earlier, a transformation of the mixture fraction in the flow field for the partially premixed flame into the mixture fraction space for the diffusion flame is required to utilize the transient flamelet libraries that are based on the diffusion flame. It is necessary for the test case computations to use this transformation, in order to predict the flame structure and the stable products. This transformation is necessary for the cases, where the flame structure is restricted by the diffusion reaction zone. In the case of a diffusion flame the mixture fraction varies between 0 and 1. In the case of a partially premixed flame the value of mixture fraction should lie between the $Z_{mj,min}$ and $Z_{mj,max}$ depending upon the boundary conditions.

However, the reactants (additional information) cannot be predicted well with this transformation. The values are extracted directly from the Burke-Shumann diagram.

The transformation procedure is explained as follows:

The flame structures that are similar in the physical space can be compared in the $Z$- space. The beginning and the end of the diffusion zone should be similar in

<table>
<thead>
<tr>
<th>Scalar</th>
<th>Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\chi$ [1/sec]</td>
<td>0.01, 0.1, 1.0, 10.0, 15.0, 30.0</td>
</tr>
<tr>
<td>$\tau$ [sec]</td>
<td>0.00001, 0.0001, 0.001, 0.01</td>
</tr>
<tr>
<td>$c$ [-]</td>
<td>0.0, 0.1, 0.2, 0.4, 0.6, 0.7, 0.8, 0.9, 1.0</td>
</tr>
</tbody>
</table>

Table 4.2: New range of the transient flamelet library
both the flame structures. This can be achieved by transforming the partially premixed flame into the $Z^*$ space normalised between 0 and 1. The stoichiometric point is kept fixed. The assumption of fixing the stoichiometric point is not absolutely accurate due to the fact that the stoichiometric point shifts with respect to the stagnation point as the partial premixing is increased.

The flamelet model is modified to account for the transformation. The flow field is firstly calculated in the normal $Z$-space. The mixture fraction field is then transformed into the $Z^*$ space before extracting the species concentration values from the transient flamelet libraries. In this way the partially premixed flame should compare reasonably well with the diffusion flame structure.

If $Z < Z_{st}$, then

$$Z' = Z_{st} \left( \frac{Z - Z_{min}}{Z_{max} - Z_{min}} \right)$$

(4.15)

In the test case, no partial premixing is carried out on the lean side boundary conditions (pure air on the lean side).

$$Z_{min} = 0$$

$$Z' = Z$$

If $Z > Z_{st}$, then

$$Z' = Z_{st} + (Z - Z_{st}) \left( \frac{1 - Z_{st}}{Z_{max} - Z_{st}} \right)$$

(4.16)

The above mentioned transformation is done in the existing flamelet code. A first attempt has been made to use the transient flamelet model for the prediction of the turbulent partially premixed flame.
Figure 4.11 Temperature profiles in the transformed mixture fraction space (based on $N_2$) for $\phi_{\text{rich}} = 2.8$, and $\phi_{\text{lean}} = 0.0$. The corresponding diffusion laminar flame is also plotted.

Figure 4.12 Species ($H_2O$) profiles in the transformed mixture fraction space (based on $N_2$) for $\phi_{\text{rich}} = 2.8$, and $\phi_{\text{lean}} = 0.0$. The corresponding diffusion laminar flame is also plotted.

The figures 4.11, 4.12 show the influence of the transformation for laminar counterflow flames. These figures represent the temperature and the product concentration profiles in the mixture fraction space, normalised between 0 and 1. The
equivalence ratio on the rich side is 2.8 and the lean side equivalence ratio is 0. It can be observed from the figures that the flame structure is not exactly the same. The maximum temperature difference on the rich mixture side is about 300°K between the transformed partially premixed flame and the diffusion flame in the mixture fraction space. It can be stated that the structure are comparable qualitatively for both the flames. This aspect is considered into account for the computation of the turbulent partially premixed flame using the transient flamelet model.
5 Test Case

The partially premixed piloted jet turbulent flame (Flame-D) [5] is computed using the transient laminar flamelet model [24]. The goal of the work is to check the capability of the transient laminar flamelet model for the prediction of the structure of partially premixed flames.

5.1 Partially Premixed Piloted Jet Flame

5.1.1 Flame Configuration

Figure 5.1 shows the sketch of the burner geometry of the piloted methane/air flame. The measurements on this flame were carried out at Sandia Laboratory [5]. Temperature and species measurements have been carried out by Barlow et al. [6]. Velocity profiles have been provided by Hassel et al. [4]. The main jet is a mixture of 25%CH₄ and 75% Air by volume. The Pilot is a lean premixture (φ = 0.77) of C₂H₂, H₂, air, CO₂ and N₂ with the same equilibrium composition and nominal enthalpy as methane/air at this equivalence ratio. The dimensions of the nozzles can be read from the sketch (cf. Figure 5.1). The details about the operating data are listed in the Table 5.1 [4].

![Sketch of the inlet nozzles of the piloted methane/air flame (Flame-D Sandia)](image-url)
### Main jet
- Inner diameter \( d \): 7.2 mm
- Wall thickness: 0.25 mm

### Pilot annulus
- Inner diameter: 7.7 mm
- Outer diameter \( D \): 18.2 mm
- Wall thickness: 0.35 mm

### Burner outer wall diameter
- 18.9 mm

### Wind tunnel exit
- 30 cm by 30 cm

### Main jet
- Velocity \( U_{\text{bulk}} \): 49.6 m/s
- Temperature: 294 K
- Pressure: 0.993 atm
- Kinematic viscosity: \( 1.58 \times 10^{-5} \) m²/s
- Reynolds number \( Re \): 22400

### Pilot jet
- Equivalence ratio \( \varphi \): 0.77
- Mixture fraction \( F_{\text{H,C}} \): 0.27
- Temperature: 1880 K
- Density: 0.1830 kg/m³

### Coflow
- Coflow velocity \( U_{\text{cf}} \): 0.9 m/s
- Temperature: 291 K
- Pressure: 0.993 atm
- Stoichiometric mixture fraction \( F_{\text{stoic}} \): 0.351

### Visible Flame Length \( L_{\text{vis}} \)
- 67d (48 cm)

**Table 5.1: Operating data for methane/air flame (Sandia flame D)**

The Raman and LIF measurements of the temperature, mixture fraction, major and minor species are available at the centerline and radial profiles at several downstream positions listed in Table 5.2.
Sandia measured a whole set of flames in a large range of Reynolds number (Re=1100, Sandia Flame A to turbulent flames with high Reynolds number close to extinction, Re=44800, Sandia Flame F) [6]. Flame-D of this set is selected as a test case for assessing the capabilities of the existing transient laminar flamelet model, due to the fact that this flame is partially premixed in nature. Moreover, this flame is stable with no visible local extinction/reignition phenomenon. So it is appropriate to test the existing model for the prediction of partially premixed turbulent flames. It is to be remembered that this model is originally developed for predicting the local extinction/reignition phenomenon in diffusion flames.

Partial Premixing with air reduces the flame length in this test flame and produces a more robust flame than pure CH₄ flame. This flame is operated at a very high Reynolds number with little or no local extinction with a pilot flame. The experimentalists believe that the flame burn as diffusion flame with no strong evidence of premixed reaction in the fuel rich CH₄/air mixtures. Our calculations show that the flame length is not predicted with the steady state laminar flamelet model, which is developed for diffusion flames. So in our opinion this flame must have few regions of local premixed combustion in the flow field, which was not noticed by the experimentalist in their experiments. On the other hand the transient laminar flamelet model with transformation of the mixture fraction in the normalised value between 0 and 1, is able to capture the flame length. The results will be discussed in the later section (see section 5.1.2).

The flow field for the present computations are solved using the commercial CFD package [62]. The standard turbulence model based on isotropic viscosity assumption (k-ε model) is used for the turbulent flow field. The Pope correction constant [78] in the ε - equation is applied to account for the jet spreading.

The computational domain consists of 120x90x3 grid nodes. It covers the axial distance from 0 till 80 main jet diameters and 0 - 20 diameters in the radial direction. Grid refinement is carried out around the centre axis of the burner to account for the boundary conditions of the inlet velocity profile.
5.1.2 Results and Discussion

For the following numerical results to be compared with the experimental data, the reaction progress variable is defined as the normalized fuel mass fraction (refer equation 2.14). We have tested the definition of the reaction progress variable based on the normalized product, namely $H_2O$ mass fraction (refer equation 2.16). Since no much difference in the flame structure is observed in the computations, so the definition of the reaction progress variable based on the fuel mass fraction is used for the present computations using the transient laminar flamelet model (TLFM).

The mixture fraction as suggested by Bilger [10] is defined in the equation 5.1. This definition is used by other modellers, who computed this flame, and the experimentalists for comparing their results.

\[
Z = \frac{(0.5(Y_H - Y_{2H}))/W_{TH} + (2(Y_C - Y_{2C}))/W_{TC}}{(0.5(Y_{1H} - Y_{2H}))/W_{TH} + (2(Y_{1C} - Y_{2C}))/W_{TC}} \tag{5.1}
\]

where

$Y_H, Y_C$ denote the local element mass fraction of hydrogen and carbon in the flow field, $Y_{1H}, Y_{1C}$ are the element mass fraction of hydrogen and carbon in main jet, $Y_{2H}, Y_{2C}$ are the corresponding element mass fractions in the coflow stream.

Figure 5.2 shows the normalized velocity profile along the axial direction. The simulated mean axial velocity along the axis predicts qualitatively the measured axial velocity profile. The differences observed lie in the prediction of the flame temperature. In the present study, the standard turbulence model from the commercial code [62] is used. The computed velocity is higher than the measured velocity in the downstream area of the simulated domain. This is due to the effect of the heat release in that region, which is caused by the higher simulated flame temperature.

Figure 5.3 shows the mixture fraction profile along the axis. It can be observed that the transient flamelet model coupled with the CFX-TASCFLOW commercial code is able to predict the mixture fraction for the partially premixed turbulent flame. The mixing is very rapid at the burner exit as compared with the measured value. This mixing is related to the use of the turbulence model. In the downstream region the mixture fraction simulated captured the measured value.

Figure 5.4 represents the temperature profile along the axial direction for the numerical and the experimental investigation of the turbulent partially premixed flame. It can be observed that the flame length is predicted well with the TLFM using new flamelet libraries. The computed peak temperature lies higher than the experimental value for the peak temperature. This is due to the radiation effects, which are not accounted in the TLFM model. The position of the peak temperature is predicted quite well with the existing flamelet model.
Fig. 5.2 Comparison of the experimental and the numerical profile of the normalized velocity along the axial direction.

Fig. 5.3 Comparison of the experimental and the numerical profile of the mixture fraction along the axial direction.
Fig. 5.4 Comparison of the experimental and the numerical profile of the temperature along the axial direction.

Fig. 5.5 Comparison of the experimental and the numerical profile of the temperature in the mixture fraction space along the axial direction.
Figure 5.5 shows again the temperature profile in the mixture fraction space. This profile indicates the difference between pure diffusion and real partially premixed flame. The transformation is carried out for $Z > Z_{st}$, such that the mixture fraction in the partially premixed flame is comparable to the pure diffusion flamelets in the libraries and corresponding values of the species are taken from these libraries. As shown in the laminar flames, the influence of the transformation is also observed in the turbulent flame tested. The temperature predicted are lower for the mixture fraction in the transformed space. So at the exit of the nozzle, the temperatures are lower causing underprediction of the axial velocity (refer Figure 5.2).

Figure 5.6 shows the products profile ($CO_2$, $H_2O$) along the axial direction. The qualitative trend is predicted by the computations. The positions of the peak concentrations for all the products species are captured in the numerical simulations.

Figure 5.7 shows the intermediate product profile ($CO$, $H_2$) along the axial direction. The $CO$ and $H_2$ is predicted only in the region, where the mixture fraction is less than 0.4 (refer Figure 5.8). This is due to the fact that the $CO$ and $H_2$ is produced in a thin reaction zone and is unable to transport. The model does not take into account the transport of the intermediate species. The increase in the scalar dissipation rate leads to incomplete combustion causing the $CO$ and $H_2$ species to be produced over the whole range of the mixture fraction. The Figures 5.10 till 5.14 show the product profiles generated in the flamelet libraries at different scalar dissipation rate. A complete combustion ($c = 1.0$) and sufficient residence time ($\tau = 0.01$ sec) is considered for the plots. The idea is to show the influence of the scalar dissipation rate and the mixture fraction on the concentration of the product profiles. In the tested partially premixed turbulent flame, the scalar dissipation rate is not extremely high along the axial direction for $x/d < 40.0$ and the mixture is very rich (mixture fraction $> 0.4$). The scalar dissipation rate is lower along the axial direction for $x/d > 40.0$. However, the mixture fraction is getting leaner along the axial direction further downstream. It can be seen from the figures showing the concentrations of the products that a certain condition for the scalar dissipation rate and the mixture fraction is required for the production of the intermediate product species over the whole range of the mixture fraction. It has been shown earlier that the models based on the flamelet assumptions [26] are unable to predict the concentration of the $CO$ species. Additional approaches may be required to predict the correct concentration of these species. The $CO$ consumption reaction is much slower as compared with the other main species reactions. There is a need of treating $CO$ species separately in the flamelet approach, e.g. by solving the transport equation for $CO$ or by considering $CO$ concentration in the initial mixing composition required for the development of the transient libraries. The existing flamelet library generation does not take into account the intermediate species in the initial mixing line for a operating condition of the reaction progress variable and the scalar dissipation rate [24]. Bajaj et al. [2] had tested the transported method for the prediction of the $CO$ concentration in the gas turbine burner and were able to get better predictions as compared with directly taking values of the $CO$ concentration from the flamelet libraries. In the existing code, another method has
to account for the CO concentration in the initial conditions for the mixing states in the development of the transient libraries.

Fig. 5.6 Comparison of the experimental and the numerical profiles of products (H$_2$O, CO$_2$) along the axial direction.

Fig. 5.7 Comparison of the experimental and the numerical profiles of reactants (CO, H$_2$) along the axial direction.
Fig. 5.8 Comparison of the experimental and the numerical profiles of reactants (CO, H₂) in the mixture fraction space along the axial direction.

Fig. 5.9 Numerical Prediction of the scalar dissipation rate along the axial direction.
Fig. 5.10 Products concentration profiles in the mixture fraction space for the transient flamelet libraries based on the diffusion flame ($c=1.0$, $\tau = 0.01$ sec, $\chi = 0.01$ [1/sec]).

Fig. 5.11 Products concentration profiles in the mixture fraction space for the transient flamelet libraries based on the diffusion flame ($c=1.0$, $\tau = 0.01$ sec, $\chi = 0.1$ [1/sec]).
Fig. 5.12 Products concentration profiles in the mixture fraction space for the transient flamelet libraries based on the diffusion flame ($c=1.0$, $\tau = 0.01$ sec, $\chi = 1.0$ [1/sec]).

Fig. 5.13 Products concentration profiles in the mixture fraction space for the transient flamelet libraries based on the diffusion flame ($c=1.0$, $\tau = 0.01$ sec, $\chi = 1.0$ [1/sec]).
Fig. 5.14. Products concentration profiles in the mixture fraction space for the transient flamelet libraries based on the diffusion flame ($c=1.0$, $t=0.01$ sec, $\chi=15.0$ [1/sec]).

Figures 5.15, 5.16 and 5.17 compare the numerical and the experimental results for the temperature, the mixture fraction and the products (CO$_2$, H$_2$O) along the radial direction at a normalized height of $x/d = 15$. The peak temperature simulated is higher than the measured values. It may be due to the radiation heat loss in the experimental flame, which is not accounted in the TLFM combustion model. The mixture fraction is predicted well in the computational results. However, a certain differences are observed at radial distance near the axis. The mixing is faster than that in the experimental flame, giving rise to lower value of the mixture fraction in the computations. The position of the peak product concentration is predicted well with the combustion model and the model is able to capture qualitatively the species profiles. However, the peak values predicted are higher due to the higher temperature simulated.

Figures 5.18, 5.19 and 5.20 show the temperature, the mixture fraction and the product (H$_2$O, CO$_2$) profiles at the normalized axial height of $x/d = 30$. The model is able to capture qualitatively the important features of the flame in the radial direction. The position of the peak temperature is predicted well. Due to the radiation effects, the peak temperature is higher than the measured peak temperature. The mixing is faster near the axis region, leading to lower value of the mixture fraction.
Fig. 5.15 Comparison of the experimental and the numerical profiles of temperature along the radial direction (at height x/d = 15).

Fig. 5.16 Comparison of the experimental and the numerical profiles of the mixture fraction along the radial direction (at x/d = 15).
Fig. 5.17 Comparison of the experimental and the numerical profiles of 
$\text{H}_2\text{O}$ and $\text{CO}_2$ along the radial direction (at height $x/d = 15$).

Fig. 5.18 Comparison of the experimental and the numerical profiles of 
temperature along the radial direction (at height $x/d = 30$).
Fig. 5.19 Comparison of the experimental and the numerical profiles of the mixture fraction along the radial direction (at x/d = 30).

Fig. 5.20 Comparison of the experimental and the numerical profiles of H₂O and CO₂ along the radial direction (at height x/d = 30).
Fig. 5.21 Comparison of the experimental and the numerical profiles of temperature along the radial direction (at height $x/d = 45$).

Fig. 5.22 Comparison of the experimental and the numerical profiles of the mixture fraction along the radial direction (at $x/d = 45$).
Fig. 5.23 Comparison of the experimental and the numerical profile of the normalized velocity along the radial direction (at $x/d = 45$).

Fig. 5.24 Comparison of the experimental and the numerical profiles of $H_2O$ and $CO_2$ along the radial direction (at height $x/d = 45$).
Figures 5.21, 5.22, 5.23 and 5.24 represent the temperature profile, the mixture fraction and the normalized velocity profile in the radial direction at $x/d = 45$. The predicted profiles and the experimental profiles are in good agreement for this height within the experimental uncertainty. The normalized velocity along the axis at this height $y/d = 45$ is higher than the experimental value. The reason may lie in the very fine grid near the burner axis and coarse grid away from the burner axis.

It has been demonstrated that the first attempt to use the existing transient flamelet model for the prediction of the turbulent partially premixed flame is successful. The potential and the weakness of the model is discussed. The carried out computation shows that the further development of the model is justified in order to predict the turbulent partially premixed flame more accurately.

### 5.1.3 Effect of Partial Premixing on the Flame Structure

The main jet stream configuration in the Flame-D is varied to observe the effect of partial premixing on the turbulent flame structure. Two new cases were computed with $\phi_{\text{main}} = 1.7$ and 2.0. The flame structures of the cases $\phi_{\text{main}} = 1.7$ and 2.0 are a single flame structure at the laminar level due to very high Reynolds number. So it is justified to compute these flames with the existing flamelet model using the newly generated libraries. The original main jet composition in the Flame-D is $\phi_{\text{main}} = 3.17$.

![Fig. 5.25 Comparison of the experimental and the numerical profile of the temperature along the axial direction (effect of equivalence ratio on the flame structure).](image-url)
Figure 5.25 shows the temperature profiles in the axial direction. It can be observed that the flame length decreases with the increase in the partial premixing in the main jet stream. These observations are similar to the observations made by other experimental groups for the laminar and turbulent coflow partially premixed flames [49]. The maximum temperatures predicted along the axis are similar for all the three computed flames.

Figure 5.26 shows the profiles of the mixture fraction along the axial direction for all the three flames. It can be observed qualitatively that the mixing decreases with the increase in the partial premixing of the main jet stream for the tested cases. The TLFM combustion model predicts qualitatively the trends that are expected by varying the partially premixed configuration in the coflow turbulent piloted jet flame.

Figure 5.27 represents the corresponding normalized velocity profiles along the axis. The effects of the heat release can be observed on the velocity profiles for the cases $\phi_{\text{main}} = 1.7$ and 2.0. The velocity profiles are the same near the nozzle exit region. This may be due to the fact that the region is outside the mixing zone and no reaction has been yet started. The gradient of the velocity profiles are stronger in the region of the reaction zone and the gradient increases with higher partial premixing. The qualitative effect of the heat release on the velocity profiles can be observed from these partial premixed flames.

![Figure 5.26](image)

**Fig. 5.26** Comparison of the experimental and the numerical profile of the mixture fraction along the axial direction (effect of equivalence ratio on the mixing).
5.1.4 Future scope of the model

In the present work a first attempt is made to use the existing model for the prediction of the partially premixed turbulent flames. Some modifications are carried out to predict the flame length correctly with the transient flamelet model. The model showed promising results for the flame length predictions and also qualitatively the concentration of the stable products (H₂O and CO₂). However, further development of the model is required in order to predict accurately the intermediate stable products, specially CO and H₂.

This model should be able to predict the turbulent partially premixed flames, provided the corresponding transient flamelet libraries are adapted for these partially premixed flames. Before starting with any computation, flamelet libraries are needed according to the boundary conditions in the turbulent reacting flow field. However, the transient flamelet library generation is computationally expensive.

It can be noticed that the temperature profile curve is not smooth from the near exit region of the nozzle till the normalized axial distance of 40 main jet diameters. The reasons lie in the selection of the pdf function for the reaction progress variable. In the model, a single - δ function is used for describing the statistical distribution of the reaction progress variable. However, it is clear from the Figure 5.28 that the variation of the reaction progress variable till x/d = 40 is quite large and the single - δ function is not suitable. The variation of the reaction progress variable for x/d > 40 is quite small. In this region the single - δ function captures the profile of the temperature smoothly. So it is suggested to use a double - δ function.
or $\beta$ - function for the pdf's of the reaction progress variable. In the premixed combustion, it is standard to use the double - $\delta$ pdf shapes for describing the fast chemistry [7].

![Graph showing fluctuations of $C$ vs $x/d_{fuel}$](image)

**Fig. 5.28** Maximum possible variation in the reaction progress variable along the axial direction.

### 5.2 Conclusions from this section

The transient flamelet model with the transformation of the mixture fraction in the normalised mixture fraction space of the diffusion flame, along with the new transient flamelet libraries is able to capture the important features of the partially premixed turbulent flame. This justifies the use of the existing TLFM code for the prediction of turbulent partially premixed flames.

The tested flamelet model uses a single-$\delta$ function for the pdf of the reaction progress variable, the scalar dissipation rate and the turbulence time scale. However, it has been shown in the computational results that the variation of the reaction progress variable is dominant in the region of the flammable mixture. This does not justify the use of a single-$\delta$ function for the reaction progress variable. It has been observed that the prediction of the intermediate species concentrations are not good by the existing transient flamelet model. The variation of the scalar dissipation rate and the mixture fraction plays an important role in this region. The CO predictions can be made better by solving the transport equation of the CO as a postprocessing and the source term for the equation can be taken from the flamelet libraries. In order to get the production rate for the CO species from the libraries, modification of the library generation procedure is required.
The effect of the partial premixing is observed in the flame structure. The flame length decreases with the increase in the partial premixing of the main jet stream. These observations of the flame length are in accordance with experimental observation by other research groups [49] on the partially premixed turbulent co-flow flames.

The peak temperature along the axis increases slightly with the increase in partial premixing for present numerical investigations.

The assessment of the existing transient flamelet model is carried out in the present study. The model shows promising results for capturing the important characteristics of the turbulent partially premixed flame. So it is justified to modify this model further to predict turbulent partially premixed flames with greater accuracy.
6 Summary

6.1 Laminar Partially Premixed Flames

6.1.1 Flame Structure

We have investigated the laminar partially premixed counterflow flames in the present study. The partially premixed flames show a distinct double flame structure under certain boundary conditions of equivalence ratios and strain rates.

The double zone structure comprises two separate reaction zones, one rich premixed and the other nonpremixed. If the fuel/air mixture at the rich boundary lies outside the flammability limit, then the mixture is only ignitable if sufficient energy and radical pool is available from the nonpremixed reaction zone. The position of the rich premixed reaction zone depends upon the energy flow from the corresponding nonpremixed zone. Reactants are consumed differently in the two zones. In the rich premixed zone, the methane/air mixture reacts to produce the partially oxidized stable species CO and H₂. These intermediates are convected towards the nonpremixed zone and oxidized by the oxygen supplied from the lean side.

It has been shown that the double zone structure helps in reducing the NO emissions as compared to a comparable nonpremixed flame. In the rich premixed reaction region little oxygen is available for NO production through the prompt mechanism. If the rich mixture equivalence ratio is greater than 1.6, then no NO is produced in that region via prompt mechanism. This leads to lower total NOx emissions from laminar flames with a double reaction zone structure as compared to nonpremixed flames.

We also observed that the stabilization of the double flame structure is very sensitive with respect to the equivalence ratio and the strain rate for a counterflow/coflow laminar flame.

6.1.2 Reaction Mechanism

The performance of several reaction mechanisms (GRI 2.11, GRI 3.0, CEC and CECR) for methane/air combustion are investigated for the prediction of the above mentioned double flame structure for laminar partially premixed flames. The CECR reaction mechanism is able to predict better the position of the rich flame in the double flame structure as compared with the predictions of the other mentioned reaction mechanisms. The sensitivity analysis is carried out for the GRI 3.0 reaction mechanism and the "suspicious" reactions are found that are causing the wrong prediction of the position of the rich premixed flame attached to the diffusion flame. The reaction rates are compared with the NIST experimental database. These rates are modified for the better prediction of the position of the rich premixed flame in the double flame structure. The modified rates are found to be lying within the experimental uncertainty. In order to justify our modi-
Summary

ification, representative experiments are required for determining the C₂ concentrations in the partially premixed laminar flames.

We also investigated the performance of the available reaction mechanisms for predicting the NO emissions in the laminar partially premixed flames. None of the mechanism is able to predict the NO concentration correctly. The modified GRI 3.0 reaction mechanism gives a better prediction of the NO concentrations as compared with the predictions of the other mechanisms in the partially premixed laminar counterflow flames. The reasons for the incorrect prediction of the NO emissions lie in the incorrect prediction of the CH concentration, which plays an important role in the initiation of prompt mechanism for NO production. However, there are very few experiments carried out in determining the CH concentration with laser diagnostics methods in the laminar rich partially premixed flames showing double flame structure. So new experiments are needed for laminar partially premixed flames to measure the CH concentration in the rich premixed region of the double flame structure. This would help to further develop these mechanisms to predict NO correctly.

6.2 Turbulent Partially Premixed Flames

The turbulent diffusion flames can be described with the two variables - namely the mixture fraction and the scalar dissipation rate - with the flamelet approach of the combustion modeling. On the other hand the structure of the premixed turbulent flames are predicted well using variables as, the reaction progress variable and the strain rate (scalar dissipation rate) in the flamelet models. So in order to describe the partially premixed turbulent flames using the fast chemistry approach, the important features of flames can be captured by determining the relationship of all the scalars as a function of the mixture fraction, the reaction progress variable, the scalar dissipation rate and flamelet evolution time.

The in-house built TLFM uses all these three variables, namely the mixture fraction, the reaction progress variable and the scalar dissipation rate, for describing the turbulent diffusion flames with very high Reynolds number. This model is used to predict the extinction/re-ignition phenomenon in the turbulent diffusion flames. The range of the reaction progress variable in order to predict the re-ignition phenomenon in the diffusion flame cases is assumed in the flamelet libraries. This range is from 0.5 till 1.0 and is able to capture the re-ignition processes.

The existing TLFM with transient laminar flamelet libraries is unable to capture the flame structure for turbulent partially premixed flames due to this insufficient range for the reaction progress variable. The new transient laminar flamelet libraries are developed covering the whole range of reaction progress variable from 0 till 1 with steps of 0.1. Moreover, the flamelet model is modified by using the transformation of the mixture fraction space into the normalised mixture fraction space (value between 0 and 1) of the diffusion flame. In this way the partially premixed flame compares reasonably well with the diffusion flame structure in the mixture fraction space. The important features of the turbulent partially premixed flame, i.e. maximum temperature, flame length, species concentrations, are captured with the existing TLFM using new transient flamelet libraries.
Summary

The existing flamelet code uses a single-δ function for the pdf of the reaction progress variable. This pdf function is not justified for the region containing the flammable mixture with large variation of the reaction progress variable. So, a double-δ function or β-function for the reaction progress variable is suggested. This is considered to be future work for the modification of the transient laminar flamelet model.

The existing combustion model is unable to predict the CO concentrations. It is due to the fact that CO is not considered in the initial mixing state for each input of the reaction progress variable. The local reaction progress variable defines the mixing composition of the partially premixed reactants and products. This assumption of having not CO in this mixing composition is not correct. Moreover, the CO production in the library generation depends strongly on the mixture fraction and the scalar dissipation rate. It is suggested to use the transport equation method for computing the concentration of the CO species. It is known that the CO chemistry is slower than the other reaction rates, so it is not correct to take the concentration of the CO species directly from flamelet libraries.

The effect of the partial premixing is also observed in the numerical calculation of the Flame-D with different equivalence ratios in the main jet stream. The flame length decreases with the increase in the partial premixing. These observations are also found in the experiments on the ELCO household burner, which was operated in the partially premixed mode.

In the present work, the assessment of the transient laminar flamelet model is done for the prediction of the turbulent partially premixed flames. This model shows promising results and can be used for the design of practical burners, operated in partially premixed modes. However, improvement of the predictive potential are needed by considering the above mentioned features.
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