I, Ahmed S AlAdawy, hereby submit this original work as part of the requirements for the degree of Doctor of Philosophy in Aerospace Engineering.

It is entitled: Effects of Turbulence on NOx Emissions from Lean Perfectly-Premixed Combustion

Student’s name: Ahmed S AlAdawy

This work and its defense approved by:

Committee chair: Jongguen Lee, Ph.D.

Committee member: Ahmed M ElKady, Ph.D.

Committee member: Bassam Sabry Mohammad Abdeenabi, Ph.D.

Committee member: Shaaban Abdallah, Ph.D.

Committee member: San-Mou Jeng, Ph.D.
Effects of Turbulence on NO$_x$ Emissions from Lean Perfectly-Premixed Combustion

A Dissertation Submitted to the
Graduate School of the University of Cincinnati
In Partial Fulfillment of the Requirements for the Degree of

DOCTOR OF PHILOSOPHY (Ph.D.)
In the Department of Aerospace Engineering and Engineering Mechanics
Of the College of Engineering and Applied Science

By
Ahmed Saad Ahmed AlAdawy
March 2014

B.S., Mechanical Power Engineering, Cairo University, Egypt
M.S., Mechanical Power Engineering, Cairo University, Egypt

Committee Chair: Prof. Jongguen Lee
Abstract

Effect of turbulence on NO$_x$ emissions is studied for a perfectly-premixed combustor running on methane/air at the mixture inlet temperature of 495 K and the combustor pressure of 5 atm over a range of equivalence ratios (0.54 - 0.85). Turbulence level is varied by a factor of 2-3 by changing the length of a portion of channels in a perforated plate on which the flame is stabilized. Simultaneous PIV and OH-PLIF measurements are used to calculate the relevant turbulence parameters such as turbulence intensity and turbulence mixing time scales, and define the flame structure such as average flame height and hence flame zone residence time scales, respectively. Effect of turbulence level is found to have negligible effect on NO$_x$ emissions level. NO$_x$ emissions are predicted by using a partially-stirred reactor (PaSR) model in CHEMKIN-PRO package with the measured turbulence parameters as the input. The modeling results agree very well with the measured NO$_x$ emission level but show that it slightly increases with the increase of turbulence level.
Acknowledgment

I would have not pursued my PhD degree without the non-intermittent guidance and supervision of Prof. Jongguen Lee, from whom I learned extensively since my first days in the program. I owe Prof. San-Mou Jeng, my first mentor at University of Cincinnati (UC), a lot since he helped me through my study, research and academic progress since the very first moment at UC. I would like to thank Prof. Shaaban Abdallah for being a wonderful teacher and a helpful consultant, who has never stopped to provide continuous fruitful advice. The faculty and staff at the Combustion Research Laboratory and UC, especially Samir Tambe, Jinkwan Song, Yi-Huan Kao, Curtis Fox and Robert Ogden, provided continuous logistical and technical assistance. I cannot thank them enough for their help, trust and support.

This dissertation would have not been possible without the help of so many people in many ways. First thanks are due to General Electric - Global Research Center (GE-GRC) for funding this project. Much gratitude is due to Dr. Bassam Abdelnabi at GE-GRC and Dr. Ahmed Elkady at GE Aviation, who both have supported my work deeply through this thesis, and have provided great moral, academic and technical support.

Gratitude is, of course, due to my professors at Cairo University, from which I developed my academic career through my bachelors, masters and my teaching and research experience. I would also like to thank my friends in Cincinnati and in my hometown in Cairo for their friendship and encouragement.

It would be impossible to say enough about my amazing parents, sister and brother, and my infinitely-supportive perfect wife. All this is meaningless without them.
The greatest gratitude, thanks and praise are to Allah, our sole God and Creator. His bounties and blessing are countless. His guidance is infinite. *Al-Hamdo-LELLAH.*
Table of Contents

Abstract .......................................................................................................................... ii
Acknowledgment .......................................................................................................... iv
List of Figures ................................................................................................................ ix
List of Tables .................................................................................................................. xiii
List of Symbols .............................................................................................................. xiv

Introduction ..................................................................................................................... 1
  1.1. Introduction ............................................................................................................. 1
  1.2. Nitrogen Oxide Formation ..................................................................................... 3
    1.2.1. Thermal (Zeldovich) NO Route ......................................................................... 3
    1.2.2. Prompt (Fenimore) NO Route .......................................................................... 4
    1.2.3. N₂O-Intermediate Route .................................................................................. 5
    1.2.4. NNH Route ...................................................................................................... 6
    1.2.5. Fuel-Bound Nitrogen Route ............................................................................ 6
  1.3. Lean Premixed Combustion ................................................................................... 7
  1.4. Significance of NO Mechanisms in Lean Premixed Combustion ......................... 8
  1.5. NOₓ Reduction ...................................................................................................... 9
  1.6. Turbulent Combustion .......................................................................................... 11
  1.7. Turbulence-Chemistry Interaction ....................................................................... 14
  1.8. Present Study ....................................................................................................... 17

Experimental Approach .................................................................................................. 19
  2.1. Introduction .......................................................................................................... 19
  2.2. Description of the Combustor ............................................................................... 19
  2.3. Emission Sampling ............................................................................................... 28
  2.4. Emission Analysis ................................................................................................. 29
    2.4.1. Fourier-Transform Infrared (FTIR) Spectroscopy ............................................. 31
    2.4.2. Paramagnetic Oxygen Analyzer .................................................................... 33
    2.4.3. Emissions Calculations and Equations ............................................................ 34
    2.4.4. Heat Loss Correction ...................................................................................... 37
4.2.3.2.  Effect of Total Residence Time ................................................................. 96
4.2.3.3.  Effect of Flame Zone Residence Time ...................................................... 99
4.3.  Carbon Monoxide (CO) Emissions ................................................................. 100
4.4.  Summary and Conclusions ............................................................................ 103

Appendix A ................................................................................................................. 105
  Flame Temperature Correction for Heat Losses ..................................................... 105

Appendix B .................................................................................................................. 109
  Dimensional Drawing for Emission Sampling Probe .............................................. 109

References .................................................................................................................. 114
List of Figures

Figure 1: Tradeoff between CO and NOx emissions, from [96] ................................................................. 2
Figure 2: Production of NO associated with the Fenimore prompt mechanism [14] ................................. 5
Figure 3: Fuel-N conversion. Solid lines denote elementary reaction pathways, while dashed arrows denote routes that involve intermediates and reactions not shown, based on modeling from [16, 19, 20]. From [21] ......................................................................................... 7
Figure 4: Combustion rig: (a) an overall view, (b) close-up at the combustor area ................................. 21
Figure 5: A sectional view of the combustor area showing main parts and measurement probes 22
Figure 6: Air inline preheater .................................................................................................................. 22
Figure 7: Fuel injection ............................................................................................................................ 23
Figure 8: Premixing of methane and air .................................................................................................. 24
Figure 9: Mixture inlet channel ............................................................................................................. 25
Figure 10: Perforated plate nozzle ......................................................................................................... 25
Figure 11: Nozzle configurations: “turbulent” 48T0110 and “normal” 48N0110 ................................. 26
Figure 12: Primary window cooling inlet ................................................................................................ 26
Figure 13: Primary window air cooling outlet ......................................................................................... 27
Figure 14: Secondary air cooling of exhaust top .................................................................................... 27
Figure 15: Emission sampling probe (a) an overall view (b) a sectional close-up view (c) positioning the emission sampling probe at 5.5” from the nozzle surface .................................................. 29
Figure 16: Schematic of the emission analysis station ........................................................................... 30
Figure 17: MultiGas FTIR unit .............................................................................................................. 32
Figure 18: FTIR Michelson interferometer ............................................................................................ 32
Figure 19: CAI 100P Paramagnetic oxygen analyzer .......................................................................... 34
Figure 20: Paramagnetic oxygen analyzer measurement principle .................................................... 34
Figure 21: Leakage closure calculation procedure ................................................................................ 36
Figure 22: Heat loss network ................................................................................................................ 37
Figure 23: Corrected temperature calculation procedure ..................................................................... 38
Figure 24: Simultaneous PIV and OH-PLIF measurement ................................................................. 40
Figure 25: PIV measurement principle ................................................................................................ 41
Figure 26: Measurement target plane ................................................................................................... 42
Figure 27: Integral length calculation from correlation parameters .................................................... 46
Figure 28: PLIF measurement principle ................................................................................................ 47
Figure 29: Extraction of flame front shape from an OH-PLIF instantaneous image .......................... 48
Figure 30: Simple chemically reacting systems; (a) constant-pressure, fixed mass; (b) constant-volume, fixed mass; (c) well-stirred reactor; (d) plug-flow reactor. [98] .................................................. 51
Figure 31: A perfectly-stirred reactor (PSR) Model .............................................................................. 52
Figure 32: Control volumes showing fluxes of mass, x-momentum, energy, and species for a plug-flow reactor (PFR). From [98].

Figure 33: Chemical reactor network (PSR+PFR) used in [82].

Figure 34: Chemical reactor network (PaSR + PFR) used in the current study.

Figure 35: A snapshot of CHEMKIN-PRO reactor network.

Figure 36: NO vs. O2-adjusted equivalence ratios for the two nozzles.

Figure 37: NO vs. O2-adjusted flame temperatures for the two nozzles (adiabatic).

Figure 38: NO vs. O2-adjusted flame temperatures for the two nozzles (with heat loss correction).

Figure 39: NO vs. O2-adjusted flame temperatures, using one curve fit.

Figure 40: NO vs. O2-adjusted Leonard and Correa’s data [81] (extrapolated to combustor pressure of 5 atm and mean residence time of 24 ms).

Figure 41: Examples of instantaneous PLIF for 48N0110, Φ = 0.60.

Figure 42: Examples of instantaneous PLIF for 48N0110, Φ = 0.66.

Figure 43: Examples of instantaneous PLIF for 48N0110, Φ = 0.78.

Figure 44: Examples of instantaneous PLIF for 48T0110, Φ = 0.60.

Figure 45: Examples of instantaneous PLIF for 48T0110, Φ = 0.66.

Figure 46: Examples of instantaneous PLIF for 48T0110, Φ = 0.78.

Figure 47: Calculation of flame height (48N0110) – (a) Φ = 0.60, (b) Φ = 0.66, (c) Φ = 0.78.

Figure 48: Calculation of flame height (48T0110) – (a) Φ = 0.60, (b) Φ = 0.66, (c) Φ = 0.78.

Figure 49: Rectangular parallelepiped flame zone.

Figure 50: PIV vectors and OH-PLIF Contours for 48N0110.

Figure 51: PIV vectors and OH-PLIF Contours for 48T0110.

Figure 52: TI for 48N0110 – Φ = 0.60.

Figure 53: TI for 48N0110 – Φ = 0.78.

Figure 54: TI for 48N0110 – cold flow.

Figure 55: TI for 48N0110 – Φ = 0.66.

Figure 56: TI for 48N0110 – Φ = 0.66.

Figure 57: TI for 48T0110 – Φ = 0.78.

Figure 58: TI for 48T0110 – cold flow.

Figure 59: TI for 48T0110 – Φ = 0.66.

Figure 60: \( \tau_t, \nu_x \) PDF for 48N0110 – Φ = 0.60.

Figure 61: \( \tau_t, \nu_y \) PDF for 48N0110 – Φ = 0.60.

Figure 62: \( \tau_t, \nu_x \) PDF for 48N0110 – Φ = 0.66.

Figure 63: \( \tau_t, \nu_y \) PDF for 48N0110 – Φ = 0.66.

Figure 64: \( \tau_t, \nu_x \) PDF for 48N0110 – Φ = 0.78.

Figure 65: \( \tau_t, \nu_y \) PDF for 48N0110 – Φ = 0.78.

Figure 66: \( \tau_t, \nu_x \) PDF for 48T0110 – Φ = 0.60.

Figure 67: \( \tau_t, \nu_y \) PDF for 48T0110 – Φ = 0.60.

Figure 68: \( \tau_t, \nu_x \) for 48T0110 – Φ = 0.66.
Figure 69: \( \mathbf{u}, \mathbf{v} \) PDF for 48T0110 – \( \Phi = 0.66 \) .........................................................82
Figure 70: \( \mathbf{u}, \mathbf{x} \) PDF for 48T0110 – \( \Phi = 0.78 \) ..........................................................82
Figure 71: \( \mathbf{u}, \mathbf{v} \) PDF for 48T0110 – \( \Phi = 0.78 \) ..........................................................82
Figure 72: Comparison of experimental and modeling results (NO\textsubscript{x} vs. O\textsubscript{2}-adjusted flame temperatures) ..........................................................84
Figure 73: Modeling results at 10 ms mean residence time compared to Leonard and Correa’s results (extrapolated to 5 atm) .............................................................................85
Figure 74: Temperature vs. total residence time for the two nozzles at \( \Phi = 0.60 \) .................87
Figure 75: Temperature vs. total residence time for the two nozzles at \( \Phi = 0.66 \) .................88
Figure 76: Temperature vs. total residence time for the two nozzles at \( \Phi = 0.78 \) .................88
Figure 77: NO\textsubscript{x} versus total residence time for the two nozzles at \( \Phi = 0.60 \) .................89
Figure 78: NO\textsubscript{x} versus total residence time for the two nozzles at \( \Phi = 0.66 \) .................89
Figure 79: NO\textsubscript{x} versus total residence time for the two nozzles at \( \Phi = 0.78 \) .................90
Figure 80: Oxygen atom levels inside the flame zone versus equivalence ratio for the two nozzles .........................................................................................................................90
Figure 81: Temperatures at end of post-flame region vs. turbulent mixing time (flame zone residence time = 3.948 ms) .................................................................................92
Figure 82: NO\textsubscript{x} at end of post-flame region vs. turbulent mixing time (flame zone residence time = 3.948 ms) .................................................................................92
Figure 83: Temperatures at the end of post-flame region vs. equivalence ratio (at flame zone residence time = 3.948 ms) .................................................................................93
Figure 84: NO\textsubscript{x} at the end of post-flame region vs. equivalence ratio (at flame zone residence time = 3.948 ms) .................................................................................93
Figure 85: Attempt for adding recirculation to the modeling chemical reactor network (using PSR for the recirculation zone) .........................................................................................96
Figure 86: Attempt for adding recirculation to the modeling chemical reactor network (using PaSR for the recirculation zone) .........................................................................................96
Figure 87: Effect of total residence time on flame temperatures for turbulent mixing time of 0.01 ms and flame zone residence time of 2.632 ms .................................................................97
Figure 88: Effect of total residence time on flame temperatures for turbulent mixing time of 0.50 ms and flame zone residence time of 2.632 ms .................................................................98
Figure 89: Effect of total residence time on NO\textsubscript{x} for turbulent mixing time of 0.01 ms and flame zone residence time of 2.632 ms .................................................................................98
Figure 90: Effect of total residence time on NO\textsubscript{x} for turbulent mixing time of 0.50 ms and flame zone residence time of 2.632 ms .................................................................................98
Figure 91: NO\textsubscript{x} vs. flame zone residence time at \( \Phi = 0.66 \) ..................................................100
Figure 92: CO level comparison between experimental and modeling results ................................101
Figure 93: Effect of turbulent mixing time on CO level at the end of the post-flame zone at flame zone residence time of 3.948 ms .................................................................................102
Figure 94: Effect of total residence time on CO level for a mixing time of 0.50 ms and flame zone residence time of 3.948 ms
List of Tables

Table 1: Flame zone residence times ..................................................................................75
Table 2: Density-weighted averages of TI .............................................................................80
Table 3: Probability density-weighted averages of mixing time scales .................................83
### List of Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>0-D</td>
<td>Zero-Dimensional</td>
</tr>
<tr>
<td>1-D</td>
<td>One-dimensional</td>
</tr>
<tr>
<td>A</td>
<td>Absorbance</td>
</tr>
<tr>
<td>A</td>
<td>Area</td>
</tr>
<tr>
<td>ATI</td>
<td>Air turbulence intensity</td>
</tr>
<tr>
<td>atm</td>
<td>Atmospheric pressure</td>
</tr>
<tr>
<td>BOOS</td>
<td>Burners Out of Service</td>
</tr>
<tr>
<td>c</td>
<td>Concentration</td>
</tr>
<tr>
<td>c&lt;sub&gt;m&lt;/sub&gt;</td>
<td>Parameter for modified Curl’s mixing model</td>
</tr>
<tr>
<td>c&lt;sub&gt;p&lt;/sub&gt;</td>
<td>Isobaric specific heat</td>
</tr>
<tr>
<td>CAEP</td>
<td>Committee on Aviation Environmental Protection</td>
</tr>
<tr>
<td>C&lt;sub&gt;2&lt;/sub&gt;H&lt;sub&gt;2&lt;/sub&gt;</td>
<td>Acetylene</td>
</tr>
<tr>
<td>CCD</td>
<td>Charge-Coupled Device</td>
</tr>
<tr>
<td>CFD</td>
<td>Computational Fluid Dynamics</td>
</tr>
<tr>
<td>CFM</td>
<td>Coherent Flamelet Model</td>
</tr>
<tr>
<td>CH</td>
<td>Carbyne radical</td>
</tr>
<tr>
<td>CH&lt;sub&gt;3&lt;/sub&gt;</td>
<td>Methyl group</td>
</tr>
<tr>
<td>CMC</td>
<td>Conditional Moment Closure</td>
</tr>
<tr>
<td>CO</td>
<td>Carbon monoxide</td>
</tr>
<tr>
<td>CO&lt;sub&gt;2&lt;/sub&gt;</td>
<td>Carbon dioxide</td>
</tr>
<tr>
<td>d, D</td>
<td>Diameter</td>
</tr>
<tr>
<td>DNS</td>
<td>Direct Numerical Simulation</td>
</tr>
<tr>
<td>E&lt;sub&gt;A&lt;/sub&gt;</td>
<td>Activation energy</td>
</tr>
<tr>
<td>EGR</td>
<td>Exhaust Gas Recirculation</td>
</tr>
<tr>
<td>FGR</td>
<td>Flue Gas Recirculation</td>
</tr>
<tr>
<td>fps</td>
<td>Frames per second</td>
</tr>
<tr>
<td>FR</td>
<td>Fuel Reburning</td>
</tr>
<tr>
<td>FTIR</td>
<td>Fourier-Transform Infrared</td>
</tr>
<tr>
<td>FWHM</td>
<td>Full Width at Half Maximum</td>
</tr>
<tr>
<td>G</td>
<td>Level-set function or Reaction-surface function</td>
</tr>
<tr>
<td>h</td>
<td>Height</td>
</tr>
<tr>
<td>H</td>
<td>Hydrogen atom</td>
</tr>
<tr>
<td>H&lt;sub&gt;2&lt;/sub&gt;O</td>
<td>Water molecule</td>
</tr>
<tr>
<td>HCN</td>
<td>Hydrogen cyanide</td>
</tr>
<tr>
<td>HiCOT</td>
<td>High-Temperature Air Combustion</td>
</tr>
<tr>
<td>HP-JSR</td>
<td>High-Pressure Jet-Stirred Reactor</td>
</tr>
<tr>
<td>ICAO</td>
<td>International Civil Aviation Organization</td>
</tr>
<tr>
<td>IEM</td>
<td>Interaction-by-Exchange-with-the-Mean</td>
</tr>
<tr>
<td>k</td>
<td>Turbulent kinetic energy</td>
</tr>
<tr>
<td>k</td>
<td>Reaction rate</td>
</tr>
<tr>
<td>l</td>
<td>Pathlength</td>
</tr>
<tr>
<td>Symbol</td>
<td>Definition</td>
</tr>
<tr>
<td>--------</td>
<td>------------</td>
</tr>
<tr>
<td>L</td>
<td>Length</td>
</tr>
<tr>
<td>$l_{t,xy}$</td>
<td>Transverse integral length scale</td>
</tr>
<tr>
<td>$l_{t,xy}$</td>
<td>Longitudinal integral length scale</td>
</tr>
<tr>
<td>LDV</td>
<td>Laser Doppler Velocimetry</td>
</tr>
<tr>
<td>LEA</td>
<td>Less Excess Air</td>
</tr>
<tr>
<td>LES</td>
<td>Large-Eddy Simulation</td>
</tr>
<tr>
<td>LMSE</td>
<td>Large-Eddy Simulation</td>
</tr>
<tr>
<td>LNB</td>
<td>Low NOx Burners</td>
</tr>
<tr>
<td>M</td>
<td>Third body</td>
</tr>
<tr>
<td>$\dot{m}$</td>
<td>Mass flow rate</td>
</tr>
<tr>
<td>$\dot{m}''$</td>
<td>Rate of mass generation</td>
</tr>
<tr>
<td>MILD</td>
<td>Moderate or Intense Low-Oxygen Dilution</td>
</tr>
<tr>
<td>MW</td>
<td>Molecular Weight</td>
</tr>
<tr>
<td>N</td>
<td>Nitrogen atom</td>
</tr>
<tr>
<td>N</td>
<td>Number of particles</td>
</tr>
<tr>
<td>N$_2$</td>
<td>Nitrogen molecule</td>
</tr>
<tr>
<td>N$_c$</td>
<td>Number of correct particles</td>
</tr>
<tr>
<td>N$_m$</td>
<td>Number of correct particles for mixing</td>
</tr>
<tr>
<td>N$_2$O</td>
<td>Nitrous oxide</td>
</tr>
<tr>
<td>NH</td>
<td>Nitrogen hydride</td>
</tr>
<tr>
<td>NH$_3$</td>
<td>Ammonia</td>
</tr>
<tr>
<td>NNH</td>
<td>Diazenyl radical</td>
</tr>
<tr>
<td>NO</td>
<td>Nitrogen oxide</td>
</tr>
<tr>
<td>NO$_2$</td>
<td>Nitrogen dioxide</td>
</tr>
<tr>
<td>NO$_x$</td>
<td>Nitrogen Oxides</td>
</tr>
<tr>
<td>O</td>
<td>Oxygen atom</td>
</tr>
<tr>
<td>OFA</td>
<td>Over-Fire Air</td>
</tr>
<tr>
<td>OH</td>
<td>Hydroxyl radical</td>
</tr>
<tr>
<td>OH-PLIF</td>
<td>Planar Laser-Induced Fluorescence using OH radicals</td>
</tr>
<tr>
<td>P</td>
<td>Pressure</td>
</tr>
<tr>
<td>P$_\Phi$</td>
<td>Joint probability density function</td>
</tr>
<tr>
<td>PaSPFR</td>
<td>Partially Stirred Plug Flow Reactor</td>
</tr>
<tr>
<td>PaSR</td>
<td>Partially-Stirred Reactor</td>
</tr>
<tr>
<td>PDF</td>
<td>Probability Density Function</td>
</tr>
<tr>
<td>PFR</td>
<td>Plug-Flow Reactor</td>
</tr>
<tr>
<td>PIV</td>
<td>Particle Image Velocimetry</td>
</tr>
<tr>
<td>PLIF</td>
<td>Planar Laser-Induced Fluorescence</td>
</tr>
<tr>
<td>ppm</td>
<td>Parts per million</td>
</tr>
<tr>
<td>psi</td>
<td>Pounds per square inch</td>
</tr>
<tr>
<td>psig</td>
<td>Pounds per square inch gauge</td>
</tr>
<tr>
<td>PSR</td>
<td>Perfectly-Stirred Reactor</td>
</tr>
<tr>
<td>$\dot{Q}$</td>
<td>Heat transfer</td>
</tr>
<tr>
<td>$R_{t,xy}$</td>
<td>Transverse correlation parameter</td>
</tr>
<tr>
<td>$R_{t,xy}$</td>
<td>Longitudinal correlation parameter</td>
</tr>
<tr>
<td>R$_u$</td>
<td>Universal gas constant</td>
</tr>
<tr>
<td>RIF</td>
<td>Representative Interactive Flamelet</td>
</tr>
</tbody>
</table>
RQL  Rich-Burn Quick-Quench Lean-Burn
SCR  Selective Catalytic Reduction
SNCR Selective Non-Catalytic Reduction
SOx  Sulphur Oxides
  t  Time
  tR Residence time
  tmix Turbulent mixing time
  T Temperature
  TI Turbulence Intensity
UHC Unburnt Hydrocarbons
  u  Transverse velocity component
v, V  Velocity
  v  Axial velocity component
  V  Volume
  Vt’ Absolute turbulence intensity
  vx  Axial velocity
  w  Circumferential velocity component
x, X  Molar fraction
X  Position
Y  Mass fraction
Z  Mixture fraction

**Greek Symbols**

ΔP  Differential Pressure
Δt  Time interval
ΔX  Displacement
ε  Turbulent dissipation
ε  Absorptvity
ρ  Density
τt,ux Transverse integral time scale
τt,uy Longitudinal integral time scale
Φ  Equivalence Ratio
ω  Mixing frequency
  \( \dot{ω}_i \) Production rate of species \( i \)

**Subscripts**

A  Flame Arrestor
cond  Conduction heat transfer perpendicular to combustor liner
c  Cooling side
conv  Convection heat transfer
cv  Control volume
eq  Chemical equilibrium
f  Forward reaction direction
g  Combustion gas side
max  Maximum
min  Minimum
mix  Mixture
rad  Radiation heat transfer
rms  Root mean square
tot  Total

**Other Notation**

\( (\_)^f \)  Variable fluctuation
\( \overline{\_} \)  Time-averaged quantity
\( (\_)^{(n)} \)  Quantity of particle \( n \)
Chapter 1

Introduction

1.1. Introduction

Protecting the environment and reducing harmful emissions has been an important issue for the last several decades, due to fast growth of civil aviation and transportation. Among those of the greatest concern are nitrogen oxides (NO$_x$), carbon dioxide (CO$_2$), carbon monoxide (CO), sulphur oxides (SO$_x$) and unburnt hydrocarbons (UHC). Some, like CO$_2$, act as greenhouse gases that can cause global warming, while others are even toxic and hazardous to the environment. Of the most significantly pollutant emissions are NO$_x$ as they can form smog and acid rain as well as affect the formation of ozone in the atmosphere. The term NO$_x$ generically refers to the sum of mono-nitrogen oxides NO and NO$_2$ (nitrogen oxide and nitrogen dioxide). NO$_x$ is produced from the reaction of nitrogen and oxygen gases through several steps mainly from combustion, especially at high temperatures. Unlike SO$_x$ which is largely unaffected by combustion reaction conditions and needs to be controlled by secondary measures, NO$_x$ may be controlled by modification of the combustion process; that has been an important incentive to study nitrogen chemistry. Despite the fact that high motor vehicle traffic produces relatively larger amounts of NO$_x$, emissions from aircraft engines are of much significant concern [1], because of their direct emission in both the upper troposphere and lower stratosphere. These emissions help in depleting the protecting layer of ozone in the stratosphere, as well as enhancing the harmful greenhouse tropospheric ozone causing problems to humans (such as respiratory diseases and infections),
animals and plants [2]. Recent studies have shown that NO\textsubscript{x} from aircraft engines lead to overall positive radiative forcing and positive potential for global warming [3-7].

Consequently, efforts have been made to restrict the increase of NO\textsubscript{x} emissions from aircraft engines. In order to meet such restrictions, the combustor designer faces either high NO\textsubscript{x} near stoichiometric and slightly-lean combustion, or CO, soot and UHC for rich combustion or high CO on the lean side, due to incomplete oxidation to CO\textsubscript{2} in low temperatures. Such trade-off (shown in Figure 1) and others like reduced combustor volumes and CO\textsubscript{2} emissions, make combustor design more challenging. Hence, better understanding of how NO\textsubscript{x} is formed and the effect of different flow conditions and combustor geometry parameters should be perceived. To have such understanding, many studies have addressed parametric investigation of different factors to improve combustor design with minimal increase of other emissions.

![Tradeoff between CO and NOx emissions](image.png)

**Figure 1**: Tradeoff between CO and NOx emissions, from [96]
1.2. Nitrogen Oxide Formation

In order to have a deep understanding of the chemistry of NO\textsubscript{x} formation, the routes by which NO\textsubscript{x} form should be understood. Five important routes have been discussed [9, 10] and studied through different literature. These routes (also called mechanisms) are the thermal (Zeldovich) NO route, the prompt (Fenimore) NO route, the N\textsubscript{2}O (N\textsubscript{2}O-intermediate) route, the NNH route, and the fuel-bound nitrogen route. After NO is formed, a small part oxidizes to NO\textsubscript{2}. However, NO\textsubscript{x} is the sum of both NO and NO\textsubscript{2}, and how NO forms is the main concern for analyzing total NO\textsubscript{x}.

1.2.1. Thermal (Zeldovich) NO Route

Thermal NO, which is also named after Zeldovich [11] who postulated this mechanism, is formed through the following elementary reactions of nitrogen (N) and oxygen (O) atoms.

\begin{align*}
O + N_2 & \rightleftharpoons NO + N \\
N + O_2 & \rightleftharpoons NO + O \\
N + OH & \rightleftharpoons NO + H
\end{align*}

The third reaction (equation (3)) completes “the extended Zeldovich mechanism”. The term “thermal” refers to the fact that the activation energy required for the first reaction (equation (1)) is high (= 319 kJ/mol), due to the strong triple bond in nitrogen (N\textsubscript{2}) molecules. This reaction can be dominant, only at high temperatures and relatively long residence times, in addition to availability of oxygen. This first reaction is the rate-limiting step of the thermal NO-formation, due to its low rate. It occurs primarily in the post-flame zone, where temperatures are typically high and residence times are fairly long.
1.2.2. Prompt (Fenimore) NO Route

Prompt NO was postulated by Fenimore [12] when he observed NO profiles measured in the post-flame gases from several hydrocarbon flames, and found out that these profiles had nonzero intercepts. This led to the nomenclature “prompt” as the NO is assumed to promptly appear. This NO could not be explained by the thermal route, due to its slow nature. Prompt NO was suggested to form because of the CH radical, which was neglected back then and considered an unimportant transient species. Under rich conditions, C₂H₂ is accumulated due to CH₃ recombination, and becomes a precursor for CH radicals, where prompt NO is favored in rich flames. At first, it was thought that CH radical would react with N₂ from air to form HCN + N, as appears in the following equation.

\[ CH + N₂ ⇌ HCN + N \] (4)

However, as reviewed in [13], disagreements led to the belief that CH reacts with N₂ to form HCN + H instead. Complex nitrogen chemistry, as shown in Figure 2, lead to the formation of NO, or N₂ which is again converted through equation (1) to NO. Prompt NO is formed at relatively low temperatures and short residence times, and is favored and dominant in the case of rich combustion.
1.2.3. N$_2$O-Intermediate Route

The N$_2$O-intermediate mechanism for NO was developed by [15]. The Intermediate N$_2$O is formed from the reaction of N$_2$ with O in the presence of a third body M, and then reacts with O to form NO as follows.

$$N_2 + O + M \rightleftharpoons N_2O + M$$  \hspace{1cm} (5)

$$N_2O + O \rightleftharpoons NO + NO$$  \hspace{1cm} (6)

$$N_2O + H \rightleftharpoons NH + NO$$  \hspace{1cm} (7)

The contribution of this mechanism is not usually significant, and hence not widely discussed, and is even sometimes ignored as a route to NO$_x$ formation. However, at lean conditions and low temperatures, thermal NO$_x$ and prompt NO$_x$ become insignificant and the N$_2$O-intermediate route becomes important to consider. Its significance is enhanced at high pressures, as stabilization of the third body M increases. Therefore, N$_2$O-intermediate route becomes a major source of NO under lean premixed combustion in gas turbines [9].
1.2.4. NNH Route

Relatively recently, this route of NO formation has been discovered [16, 17]. The initiating step is the formation of NNH from H and N$_2$.

\[ H + N _2 \rightleftharpoons NNH \] (8)

At high temperatures, equilibrium is reached rapidly. Because of its thermal instability and short lifetime, NNH is mostly converted back to N$_2$ by the backward direction of equation (8). A remaining small fraction of NNH reacts rapidly with O atoms as follows.

\[ NNH + O \rightleftharpoons NH + NO \] (9)
\[ NNH + O \rightleftharpoons N_2O + H \] (10)

Both NH and N$_2$O may be then oxidized to NO. Under certain conditions, this route may contribute significantly, as on the fuel-rich side of sheets of diffusion flame where NNH forms and reacts with O inside the flame sheet [18].

1.2.5. Fuel-Bound Nitrogen Route

Unlike the previous NO routes, that rely on nitrogen in combustion air, this route is based on small percentages of chemically-bound nitrogen in fuels such as coal or biomass. This fuel-bound nitrogen leads to formation of NH$_3$ and HCN through a sequence of very fast reactions, and N atoms eventually form to react with OH and O$_2$, forming NO$_x$. Reactions competing for N-atoms such as:

\[ N + OH \rightleftharpoons NO + H \] (11)
\[ N + NO \rightleftharpoons N_2 + O \] (12)

are the rate-limiting steps, as they are significantly slower than those forming NH$_3$ and HCN. Detailed chemistry is shown in Figure 3.
It should be noted that the formation of NO\textsubscript{x} is relatively slow, so it is mainly governed by chemical kinetics. Chemical equilibrium is not a good assumption for the calculation of NO\textsubscript{x}, though it can be used for the prediction of other emissions like CO\textsubscript{2}, CO, O\textsubscript{2} and H\textsubscript{2}O since they are much faster to form than NO\textsubscript{x}. Equilibrium NO\textsubscript{x} is about 2000 parts-per-million (ppm) while the limits are around tens of ppm. Thorough studies of nitrogen chemistry can be found in literature, e.g. [10, 14, 21, 22].

1.3. **Lean Premixed Combustion**

One of the most important factors that affect NO\textsubscript{x} production is high temperature. The dependence of NO\textsubscript{x} formation on temperature and the related chemical reactions are due to the extended Zeldovich mechanism. As can be obtained from equilibrium calculations, the highest temperature of combustion occurs at the stoichiometric ratio (the exact ratio that allows the combustion reaction with no excess of fuel or oxidizer).

To avoid NO\textsubscript{x} production at that stoichiometric ratio, either rich (excess fuel) or lean
(excess oxidizer) burning can be followed. At equivalence ratios (ratio of actual-to-stoichiometric fuel-to-air ratios) higher than unity, i.e. rich, the high-temperature peak can be avoided at the expense of high soot, UHC and CO, as well high cost of fuel. On the other hand, lean-burn engines use excess air to avoid the stoichiometric high temperatures, which is more cost efficient, and minimizes the extra emissions. Importantly enough, leaning the flame does not reduce the thermodynamic efficiency of gas turbine cycles [23].

An important addition to lean combustion is involving a premixed fuel-air supply. Lean premixed combustion not only avoids the production of NO$_x$ at stoichiometric overall equivalence ratios, but also avoids the formation of hot spots due to local stoichiometric flame zones, hence minimizes the local occurrence of high temperature and high NO$_x$.

1.4. Significance of NO Mechanisms in Lean Premixed Combustion

Considerable concern has been directed to lean premixed combustion, and through different studies, NO$_x$ mechanisms have been examined through experimental testing or chemical kinetic modeling. For instance, Corr et al.[24] used ethylene and methane as fuels in both premixed and non-premixed modes through a jet-stirred reactor. As expected, the premixed mode produced about half those from the non-premixed mode. Emissions from methane were about 25% less than those from ethylene. Prompt NO, predicted by extrapolation of Fenimore’s dependence functions, was found to be the predominant mechanism for NO$_x$ formation, although it is associated mainly with rich combustion. The N$_2$O-intermediate mechanism was also found to contribute a small amount to lean premixed combustion.

More focus on the N$_2$O-intermediate route was dedicated by Nicol et al. [25] as they studied the contribution of N$_2$O route to NO$_x$ formation in lean premixed combustion. Using a
methane fuel and chemical kinetic modeling, the three most common NOx mechanisms (thermal, prompt and N2O-intermediate) are studied by the chemical reactor model. It was found that N2O-intermediate route contribution can be 20% to 35% of total NOx at intermediate pressures (10 atm), and increases to be 40% to 45% of total NOx at higher pressures (30 atm). N2O-intermediate route contribution increases to almost 100% when total NOx emissions decrease to less than 10 ppm. As pressures decrease towards atmospheric pressure, all three main paths are considerable. Pressure dependence of NOx is relatively weak at 10 atm and increases at 30 atm to reach an approximately root pressure dependence.

Using a methane lean premixed high-pressure jet-stirred reactor (HP-JSR), modeled by appropriate chemical kinetic reactors, Rutar et al. [26] studied the contribution of NOx routes and proved the significance of N2O-intermediate NOx in lean premixed combustion. However, for equivalence ratios larger than 0.7, the significance of the prompt NOx route was found to increase, as the flame becomes relatively richer.

Hence the significance of N2O-intermediate route and prompt NO route depends on how lean the pre-mixture is. The leaner it is, the more significant the N2O-intermediate route will be.

1.5. NOx Reduction

After establishing some understanding of the relative contribution of NOx mechanisms, several ways to reduce NOx have been studied through extensive literature along the past few decades. Extensive literature of research studies has been performed to improve such methods and increase their efficiency. Among the known NOx reduction methods are:

1. Reducing Peak Temperature
   a. Flue Gas Recirculation (FGR)
b. Natural Gas Re-burning

c. Low NO\textsubscript{x} Burners (LNB)

d. Combustion Optimization

e. Burners Out of Service (BOOS)

f. Less Excess Air (LEA)

g. Inject Water or Steam

h. Over-Fire Air (OFA)

i. Air Staging

j. Reduced Air Preheat

k. Catalytic Combustion

l. Rich-Burn Quick-Quench Lean-Burn (RQL) Combustion

2. Reducing Residence Time At Peak Temperature

a. Injecting Air

b. Injecting Fuel

c. Injecting Steam

3. Chemical Reduction Of NO\textsubscript{x}

a. Fuel Re-burning (FR)

b. Low NO\textsubscript{x} Burners (LNB)

c. Selective Catalytic Reduction (SCR)

d. Selective Non-Catalytic Reduction (SNCR)

4. Oxidation Of NO\textsubscript{x} With Subsequent Absorption

a. Non-Thermal Plasma Reactor

b. Inject Oxidant
5. Removal Of Nitrogen
   a. Oxygen Instead of Air
   b. Ultra-Low Nitrogen Fuel
6. Using A Sorbent
   a. Sorbent In Combustion Chambers
   b. Sorbent In Ducts
7. Combinations Of These Methods
   One of the most important, yet challenging methods of reducing NO\textsubscript{x} is by altering the turbulence intensities involved in combustion flows. A major complexity of this method refers to the fact that turbulence is not yet fully understood. Many efforts to understand flow turbulence have been made and approximations are still made. For instance, turbulence models have been used to “close” the problem of relating turbulent flow parameters, and hence are called closure models, and vary in complexity, number of equations used and hence relative accuracy. One more challenge is providing the computation resources (and hence expense) involved with turbulence modeling. As efforts are made to make better approximation of the turbulence problem, the number of closure equations increases, and use of finer grids and smaller time steps is required. That increases expenses to a degree where compromise has to be made for efficient studies.

1.6. **Turbulent Combustion**

   Adding insult to injury, involving turbulence in combustion is much more complex, since chemical reactions interfere with turbulence and more phenomena have to be understood. Turbulence-chemistry interactions even vary in different combustion modes and with changing operating and boundary conditions. In addition, more complexities may evolve, such as
 thermoacoustic effects, as acoustic waves can mutually interact with turbulence and chemistry, which further increases challenges. Other challenges combined with turbulent combustion are unmixedness of reactants, spray atomization and vaporization in liquid fuels, and autoignition. Due to such complexity, it is difficult to generalize or sort out relations between studies of turbulent combustion in different modes and applications.

To understand turbulent combustion, various approaches can be found in different combustion books and literature [28-31]. These approaches [32] are:

1. Phenomenological approaches: by considering the phenomenon of turbulent combustion itself, independent of the underlying conservation equations.
   a. Zero-Dimensional (Quasi-dimensional): [33, 34].
   b. Age Theories (by including residence times as random variables, and working with ordinary-differential equations and chemical reactors): [35].
   c. Multidimensional Age Theories (by adding space variables to the time variables): [36, 37].
   d. Linear-Eddy Concept (one dimensional turbulence), [38-47].

2. Fluid-based approaches, where governing fluid and chemical reaction equations are studied, with averaging as necessary:
   a. Direct Numerical Simulation (DNS) which is often limited by computer resources and simulation time. [48, 49]
   b. Large-Eddy Simulation (LES), by introducing subgrid modeling to DNS [50-52]
   c. Turbulence closure equations, including 1st order algebraic, Reynolds Stress, kinetic energy (k) - dissipation (ε) and others.
3. Perturbation-based approaches (where perturbation theory is applied to reacting Navier-Stokes equations)
   a. Low-intensity, large-scale perturbation [53, 54]
   b. G-equation moment modeling, where premixed combustion is addressed in the limit in which the laminar flame thickness becomes small, where G is the level-set function, [31, 55-58]
   c. Representative Interactive Flamelet (RIF) Libraries for turbulent diffusion flames, [59] or Flame Surface Evolution Modeling (Coherent Flamelet Model (CFM)) for turbulent premixed flames [60-62]

4. Probability-density function (PDF) evolution approaches (where the randomness of turbulent statistics are imposed on the Navier-Stokes equations)
   a. Linear Mean-Square Estimation (Interaction by Exchange with Mean, IEM), [63]
   b. Coalescence-Dispersion [64]
   c. Mapping Closure [65]
   d. Euclidean Minimum Spanning Tree [66]

5. Conditioned PDF (by conditioning or assuming forms of the PDF to achieve better simplification)
   a. P(Z) for diffusion flames, where Z is the mixture fraction, [67, 68]
   b. P(c) for premixed flames, where c is the reaction progress variable [69-72]
   c. P(G) for premixed flames, where G is the level-set or reaction-surface function, [31]
   d. Conditional moment closure (CMC), [73-75]

6. Others for premixed turbulent flames
a. Renormalization of G-equation [76, 77]

b. Fractals [78, 79]

c. Pseudosolitons [80]

To stabilize turbulent flames, many methods have been applied for anchoring the combustion flame. Of the most important is stabilization over a bluff body. One example of bluff-body stabilization is using porous plates, where the reactant pre-mixture flows in jets through drilled holes in a flat plate, with the rest of the plate acting as the anchoring bluff body. Lean premixed flames stabilized by such method have been studied for instance by Leonard and Correa [81], Elkady et al. [82], York et al. [83], Wang et al. [84], and Boushaki and Sautet [85]. The work of Leonard and Correa [81] was used for validation of the present study experimental results and modeling predictions.

1.7. Turbulence-Chemistry Interaction

As previously-stated, among the efforts to limit NO\textsubscript{x} emissions is to change the velocity field of the combustion zone, an effective way is to vary the flow turbulence, or available mixing time. When flow turbulence is increased, it is often believed, at least for non-premixed flames, that nitrogen oxides can be reduced. However, a generalized overview of the effect of turbulence on NO\textsubscript{x} and chemistry-turbulence interactions needs careful investigation and thorough study of different NO\textsubscript{x} formation mechanisms and requires the monitoring of different chemical and turbulence time scales. According to the combustion mode and operating conditions, turbulence can have different effects on NO\textsubscript{x} formation. For non-premixed combustion, turbulence enhances the premixing of reactants and hence reduces the potential for hot spot formation and high flame temperatures. Accordingly, it is often established that more intense turbulence reduces NO\textsubscript{x} due
Kobayashi et al. [86] investigated the effect of inlet air turbulence on flame structure and the resulting NO\textsubscript{x} concentrations in High-Temperature Air Combustion (HiCOT) non-premixed flames. They used perforated plates of different hole diameters (keeping a constant excess air ratios) to induce different inlet air turbulence intensities. Inlet air was preheated up to 1300 K and diluted with nitrogen to reduce oxygen concentrations and realize HiCOT conditions, and propane was used as a fuel. Increasing inlet air turbulence, a shorter much lifted flame with eliminated soot radiation was observed and output NO\textsubscript{x} was significantly reduced. Although flame lifting was a reason for reducing NO\textsubscript{x}, it was also reduced for unlifted flames.

Using a finite-volume method with realizable k-\varepsilon turbulence and \beta-PDF combustion models, Hashemi et al. [87] studied the effect of air turbulence intensity (ATI) on NO formation for fuels of different hydrogen-hydrocarbon mixture fractions (100/0, 70/30, 10/90 and 0/100 H\textsubscript{2}/CH\textsubscript{4} percentages) in non-premixed combustion. They showed that increasing air turbulence decreases NO concentration in the flame zone and at the combustor outlet. The effect was observed for all different composite fuels, however enhanced most for pure hydrogen fuel. It was also obvious that increased air turbulence reduced the peak temperature of the flame, a potential for reduction of NO concentrations. Increasing the ATI led to more blending of burnt and unburnt gases, which similarly to EGR mechanism, reduces flame peak temperature due to high heat capacity of burnt gases.

Hwang et al. [88] studied the effect of turbulence intensity of inlet air on NO\textsubscript{x} emissions in H\textsubscript{2}/air non-premixed flames. They found that increasing turbulence intensity caused a significant reduction (by 72%) of NO\textsubscript{x}, especially in the condition of higher flame temperatures.
On the contrary, in premixed flames, increasing turbulence seems to either increase NO\textsubscript{x} or have a minimal effect, for the same unburnt flow velocity. Since the reactants (fuel + oxidizer) are already premixed, turbulence is usually referred to as in-combustor stirring which enhances the mixing of reactants with products. Increasing the frequency of in-combustor stirring can increase NO\textsubscript{x} levels due to different factors including activity of oxygen and hydroxyl radicals, as noted in different literature (e.g., [89]).

Through a partially-stirred reactor (PaSR) model, Correa [89] computed the effect of turbulence on NO, using 50% CO / 50% H\textsubscript{2} (by volume) fuel, premixed with air. As the mixing frequency \( \omega \) (and hence turbulence level) was increased, NO was shown to increase by a factor of 2.5 as the mixing time became small enough to affect the concentration of OH radicals. As temperature decreased by increasing mixing frequency, the reduction of NO\textsubscript{x} was attributed to oxygen behavior. The scheme included thermal and N\textsubscript{2}O-intermediate NO\textsubscript{x}, which is a subset of a complete methane scheme, studied by Correa and Braaten [90] in a methane premixed flame simulation.

For 30 atm, 1200-K inlet temperature, a full scheme for methane combustion was used and a similar PaSR was implemented to reveal that NO increased significantly by increasing frequency of in-combustor turbulence. However, the increase of NO\textsubscript{x} was attributed to both increase of flame peak temperature and change of radical paths and concentrations, in contrary to the CO/H\textsubscript{2} case [89] where temperature decreased with turbulent mixing and NO\textsubscript{x} increase was attributed to oxygen behavior.

In addition, to study the effect of turbulent mixing intensity on moderate or intense low-oxygen dilution (MILD) combustion and accompanying NO\textsubscript{x} emissions, Hamdi et al. [91, 92]
divided the flameless combustion zone into a first combustor, modeled by a perfectly-stirred reactor (PSR) and a second combustor, modeled by a PaSR. They used a GRI-Mech 3.0 scheme and found out that increasing the turbulence intensity slightly increases or decreases NO$_x$, depending on the range of mixing time scales. Decreasing the mixing time scale from 50 ms to 25 ms slightly reduced NO$_x$ while decreasing it further to the PSR limit (0 ms) slightly increased NO$_x$.

Kraft et al. [93] also developed a partially stirred plug flow reactor (PaSPFR) model to study the effect of varying turbulent mixing intensity on NO$_x$ formation from simultaneous combustion and mixing of two separate flows, one contains hot combustion products of a previous lean combustor and the other comprises additional fuel. The NO$_x$ formation is observed to shift between different (prompt, thermal and N$_2$O pathway) mechanisms. However, the overall NO$_x$ emissions slightly increase with more intense mixing.

### 1.8. Present Study

At the most of the author’s knowledge, no literature has been done elsewhere on the effect of turbulence mixing on NO$_x$ emissions from lean perfectly premixed jet flames from porous plates with accompanying chemical kinetics network models. The effect of turbulence on NO$_x$ here is studied both experimentally and using a chemical kinetics model using methane-air pre-mixture at a preheated temperature of about 495 K. Here, the objective is to analyze the effect of turbulence on lean perfectly-premixed combustion stabilized over a porous plate.

Throughout this study, the reactants are provided through two different nozzles (porous plates). Both provide the same mean flow velocity but different turbulence levels. First, an experimental rig was built to provide the flow and boundary conditions (5 atm and around 495 K
preheat). NO\textsubscript{x} and other emissions are measured for both nozzles, through a range of equivalence ratios, using a Fourier-transform infrared (FTIR) spectroscopy analyzer and a paramagnetic oxygen analyzer. Velocity field and flame structure are visualized using simultaneous particle image velocimetry (PIV) and planar laser-induced fluorescence using OH radicals (OH-PLIF).

The second part of the study uses chemical kinetics to model the NO\textsubscript{x} emissions from both nozzles, using a partially-stirred reactor (PaSR) and a plug-flow reactor (PFR). CHEMKIN-PRO code was used for the numerical computations for this reactor network. Lastly, results from both numerical and experimental approaches are presented and compared, and validated with experimental findings from previous literature [81] and conclusions are drawn. Other comments and future recommendations are suggested.
Chapter 2

Experimental Approach

2.1. Introduction

In this chapter, experimental procedure and apparatus for studying NO\textsubscript{x} emissions is presented. First, the combustor used for lean premixed combustion is illustrated and the needed procedure is explained. The measuring instrumentation to monitor NO\textsubscript{x} levels as well as other important emissions is explained. Laser diagnostics (Simultaneous PIV and OH-PLIF) used to characterize the flow and flame structure and calculate turbulence parameters are described.

In order to study the effect of turbulence on NO\textsubscript{x} emissions, two different configurations for changing turbulence levels, by changing geometry, are discussed. In order to calculate turbulence levels and assess time and length scales, PIV technique and accompanying correlations and calculations are presented. OH-PLIF technique is also discussed and explained.

Furthermore, to explain the effect of turbulence on NO\textsubscript{x} levels, chemical kinetics modeling is performed to simulate the combustion process and the linked reactions, through CHEM KIN-PRO, as will be discussed later in the next chapter.

2.2. Description of the Combustor

An overall view of the combustor rig and its different components is shown in Figure 4a, and a close-up to the main combustor area is shown in Figure 4b. A sectional view is also shown
in Figure 5.

To obtain a perfectly-premixed mixture of fuel and air at the operating conditions (5 atm pressure and 495 K inlet temperature), air is first preheated using an exit-temperature inline heater (OSRAM Sylvania 038825, 6.0 kW) to a temperature of 495 K, as shown in Figure 6). CP-grade 2.0 (99.0% pure) methane is injected radially from a ¼” tube, through 8 circumferentially-distributed holes into the preheated air pipe, as illustrated in Figure 7, to obtain a range of equivalence ratios (0.54 - 0.85). To get perfect premixing, the fuel/air mixture passes through a coiled tube bundle of length-to-diameter ratio of 280, as shown in Figure 8, and enters the inlet channel, Figure 9, where a ceramic honeycomb mesh (with length-to-diameter ratio >> 1) is used to minimize lateral flow turbulence and eddies. The air/fuel pre-mixture is then introduced to the combustor, through the perforated plate (referred to later on as “the nozzle”), illustrated in Figure 10. The nozzle is composed of a center pilot ignition hole (0.250” diameter), surrounded by 48 main pre-mixture holes, arranged in a square array with an equal spacing of 0.250” (0.635 cm) in each direction. The main diameter of each of the main holes is 0.11” (0.279 cm) and the total length is 0.875” (2.22 cm). The first part of the nozzle is composed of a flame arrestor of 0.065” diameter to prevent flashback.
Having the same total nozzle length, two different nozzle geometries (48N0110 and 48T0110) feature flame arrestor lengths of 0.25” (0.635 cm) and 0.625” (1.588 cm), respectively and are shown in Figure 11. The two nozzles are used to produce two sets of results at different turbulence intensity levels by changing the arrestor length. Turbulence level, as shown later in PIV results, is proven to have increased from the “normal” nozzle to the “turbulent” one.
Figure 5: A sectional view of the combustor area showing main parts and measurement probes

Figure 6: Air inline preheater
Combustion of the pre-mixture is initiated by a hydrogen pilot flame ignited by a voltage-difference-driven electrode, and is obtained at the surface of the nozzle. A globe flow valve is controlled at the exhaust of the combustor to keep combustion pressure at 5 atm. A pressure vessel surrounds the combustor and contains cooling air at a pressure nominally equal to the same combustor pressure and absolute difference is no more than 0.1 psi, to reduce the pressure difference on the combustor walls and hence minimize leakage potential.

In order to monitor flow conditions, different measurements are reported at all parametric studies. Pressure taps are used to convey the inlet pressure ($P_1$), combustor pressure ($P_2$) and cooling vessel pressure ($P_3$) to different pressure gauges. Differential pressure around the nozzle ($\Delta P_{\text{nozzle}}$) and around the cooling air inlet ($\Delta P_{\text{cooling}}$) are used for double-check calibration of main air and cooling air flow rates, respectively. Pre-mixture temperature at the inlet channel ($T_1$),
Combustor liner temperature ($T_2$), and cooling stream exhaust temperature ($T_3$) are recorded through K-type thermocouples. Additional measurements are monitored to protect exhaust valve hardware from thermal stresses. Cooling air exhaust temperature ($T_3$) is used for the estimation of heat losses from the combustor by convection which, as discussed in later sections, is used in the heat loss model. Different measurement ports are shown in Figure 5. Main stream (air and fuel) and cooling flow rates are measured using both Coriolis flow meters and laminar-flow-element flow meters.

Figure 8: Premixing of methane and air
Figure 9: Mixture inlet channel

Figure 10: Perforated plate nozzle
Figure 11: Nozzle configurations: “turbulent” 48T0110 and “normal” 48N0110

Figure 12: Primary window cooling inlet
Figure 13: Primary window air cooling outlet

Figure 14: Secondary air cooling of exhaust top
2.3. Emission Sampling

To draw a representative local sample from the combustion products for combustion product emission measurements, an emission sampling probe, which is basically a ⅛” stainless steel tube through which a fraction of the combustion products are extracted from the combustor. Choice of stainless steel for the emission sampling probe favors the catalysis of free radical recombination reactions [94]. In order to avoid overheating, the ⅛” extraction tube is concentrically aligned with two larger stainless steel tubes, ¼” and ⅜” in diameter and welded in such a way to create a water convection cooling cycle around the extraction tube. At a height of 5.5” above the nozzle surface, the emission sampling probe is fixed along the centerline of the combustor. The emission sampling probe is shown in Figure 15.

After exiting the combustor, the emission sampling probe is connected to heated tubing, through which the sample is electrically heated to keep the emission sample at a temperature of 150°C to avoid condensation of water vapor, so that composition of the sample is not affected. The sample is conveyed through the heated tubing to the gas emissions analysis station. The gas sampling system was frequently tested for crossover sample/coolant leakage by pressurizing water inside the cooling system, closing the cycle and observing the internal pressure over time. Pressure was observed to not drop more than 2 psi for an initial internal pressure of 120 psig, over an interval of two hours.
2.4. **Emission Analysis**

As shown in Figure 16, the gas emission analysis station begins with a sample conditioning unit, through which the sample is filtered, heated and pumped to a chilled tube bundle, referred to as the condensation (dehumidification) unit. Moisture in the sample is captured through the condensation unit, to prevent the alteration of NO concentration due to water content, and then the dry emission sample is distributed to both a paramagnetic oxygen analyzer and a Fourier-transform infrared spectroscopy analyzer.
Figure 16: Schematic of the emission analysis station
2.4.1. Fourier-Transform Infrared (FTIR) Spectroscopy

A Fourier-transform infrared (FTIR) spectroscopy analyzer (MKS MultiGas™ 2030, Figure 17) was used for the dry analysis of NO (ppm), NO\textsubscript{2} (ppm), CO (ppm) and CO\textsubscript{2} (%) emissions. The basic principle [95] of the FTIR depends on the fact that different compounds, if subjected to infrared radiation, absorb different proportions at different frequencies. The absorbance of infrared radiation by different compounds is unique, which makes structure of chemical bonds (or functional groups) distinguishable from the analysis of infrared spectra by observing peaks at which absorption occurs and their locations (corresponding wavenumbers or wavelengths). Michelson interferometer, as shown in Figure 18, is the basic component of FTIR instruments and consists of infrared light band source, a beam splitter, a fixed mirror and a moving mirror. Infrared light band splits into a beam that falls on the stationary mirror, and another beam falling on the moving mirror. After reflection at the mirror, the beams merge again to pass through the gas sample to the detector. As the moving mirror scans through different positions, the path difference between the two beams changes and interferograms can be plotted by measuring intensities at the detector versus optical path difference. Fourier Transforms are applied later to the interferograms give spectrums. Concentration of different emissions in the sample can be calculated from absorbance in infrared radiation mainly from Beer’s law:

\[ A = \varepsilon \l c \]  

where:

\( A \) = Absorbance \\
\( \varepsilon \) = Absorptivity \\
\( l \) = Pathlength
\[ c = \text{Concentration} \]

Figure 17: MultiGas FTIR unit\(^1\)

Figure 18: FTIR Michelson interferometer\(^2\)

\(^1\) [Image 1](http://img.directindustry.com/images_di/photo-g/ft-ir-multi-gas-analyzers-7020-2284991.jpg)
\(^2\) [Image 2](http://upload.wikimedia.org/wikipedia/commons/thumb/a/a1/FTIR_Interferometer.png/495px-FTIR_Interferometer.png)
Among the limitations of infrared spectroscopy is that it cannot be used for atoms, monatomic ions and noble gases due to lack of chemical bonds, as well as for homonuclear diatomic molecules such as \( \text{O}_2 \) and \( \text{N}_2 \) as they don’t possess infrared spectra due to their symmetry. Due to solubility of water with infrared transparent gas cell materials and windows and its strong infrared absorption, the gas detection cell is kept at a temperature of 150°C to eliminate doubt of condensation of water vapor to protect the FTIR windows. Since dry analysis in this present study is performed, moisture is extracted before FTIR measurement and exposure of FTIR components to moisture is minimized. MultiGas™ software (MG2000, version 06.31.06) was used for the FTIR quantitative analysis of emissions. Two calibration gas cylinders were used to check the performance of the FTIR, and recalibrate the instrument if needed. The calibration gas check was performed before and after every series of emission analysis runs. Measurements were taken after a steady state is reached in the combustor, after two minutes of changing the conditions to the following operating point.

2.4.2. Paramagnetic Oxygen Analyzer

As it is not possible to measure oxygen percentages using FTIR, a paramagnetic oxygen analyzer (CAI 100P, Figure 19) was used. It is capable of measuring dry \( \text{O}_2 \) (%) molar fractions by measuring the paramagnetic susceptibility of the sample gas by a magnetodynamic type measuring cell. The measuring cell consists of a dumbbell of diamagnetic material, which is temperature-controlled electronically at 50°C. The higher the oxygen concentration, the greater the dumbbell is deflected from its real position, detected by an amplified optical system. To return the dumbbell to its original position, current passes through a coil of wire surrounding the dumbbell, and is linearly proportional to the percent oxygen concentration in the sample gas. A schematic of the principle of operation is illustrated in Figure 20.
2.4.3. Emissions Calculations and Equations

To ensure minimum leakage from or into the combustor, a leakage closure term was calculated at all operating points. The leakage closure term checks the difference between the output (O₂ or CO₂) emissions and the estimated corresponding emissions from the combustion equation, equation (14) in case of methane fuel.

---

3 From paramagnetic oxygen analyzer manual
4 From paramagnetic oxygen analyzer manual
\[ \text{CH}_4 + \frac{2}{\phi} (O_2 + 3.76N_2) \]

\[ \rightarrow \alpha \text{CO}_2 + \beta \text{CO} + \gamma \text{O}_2 + \delta \text{H}_2\text{O} + \varepsilon \text{NO} + \eta \text{NO}_2 + \cdots + \psi N_2 \]  

(14)

where \( \phi \) is the equivalence ratio

\( \alpha, \beta, \gamma, \delta, \varepsilon, \eta, \ldots, \psi \) are the number of moles of each of the products assuming one mole of input methane

The closure may be then calculated from either \( \text{O}_2 \) emissions or \( \text{CO}_2 \) emissions, defined by the ratio of the equivalence ratio that gives the same output \( \text{O}_2 \) (or \( \text{CO}_2 \)) percentage from the equilibrium calculations, to the metered flow equivalence ratio. \( \text{O}_2 \) leakage closure may be calculated from equation (15). Similarly, The \( \text{CO}_2 \) leakage closure is calculated from equation (16).

\[ O_2 \text{ Closure} = \frac{\phi_{\text{based on } O_2}}{\phi_{\text{metered flow}}} \]  

(15)

where \( \phi_{\text{metered flow}} \) is the equivalence ratio calculated from the metered air and fuel flow rates

\( \phi_{\text{based on } O_2} \) is the equivalence ratio, back-calculated from chemical equilibrium calculations using the measured output \( \text{O}_2 \) emissions as a given input.

\[ \text{CO}_2 \text{ Closure} = \frac{\phi_{\text{based on } \text{CO}_2}}{\phi_{\text{metered flow}}} \]  

(16)

where \( \phi_{\text{based on } \text{CO}_2} \) is the equivalence ratio, back-calculated from chemical equilibrium calculations using the measured output \( \text{CO}_2 \) emissions as a given input.

The procedure followed to calculate leakage closures is summarized in Figure 21. \( \text{O}_2 \) and \( \text{CO}_2 \) leakage closures at all operating points were always above 95%.
To compare dry measured mole fractions to mole fractions calculated from chemical equilibrium calculations, conversion between wet and dry mole fractions was performed as shown in equation (17).

$$X_{i,dry} = X_{i,wet} \cdot \frac{1}{1 - X_{H_2O,wet}}$$

where $X_{i,dry}$ is the dry mole fraction of component $i$

$X_{i,wet}$ is the wet mole fraction of component $i$

$X_{H_2O,wet}$ is the wet mole fraction of water vapor, and by definition, $X_{H_2O,dry} = 0$

**Figure 21: Leakage closure calculation procedure**

Combustion efficiency was also calculated at different operating points, defined as the molar fraction of CO$_2$ emissions in the combustion products, divided by the sum of molar fractions of product CO$_2$, CO and unburnt hydrocarbons. Combustion efficiencies were always larger than 95%.
2.4.4. Heat Loss Correction

Because the combustor is externally cooled by compressed air in the pressure vessel, heat losses are carried away by different modes of heat transfer. Heat losses ($Q_{\text{loss}}$) can be calculated from internal convection, flame radiation, external convection, external radiation and conduction. Appropriate method of heat loss calculation was adopted from [96]. Heat loss model, as shown in Figure 22, takes into consideration radiation from the flame and combustion gases to the inner surface of the combustor liner ($Q_{\text{rad},g}$), internal convection heat transfer from hot combustion gases to outside the combustor ($Q_{\text{conv},g}$), conduction through the combustor liner perpendicular to the flow direction ($Q_{\text{cond}}$), external convection heat transfer carried by cooling air in the pressure vessel ($Q_{\text{conv},c}$), and external radiation from the outer surface of the combustor liner ($Q_{\text{rad},c}$). Conduction along the combustor liner was neglected as it is a very small fraction of the other heat transfer modes.

![Figure 22: Heat loss network](image)

Neglecting conduction along the combustor, energy balance from the thermal network can be established as shown in Figure 22 and equation (18) can be formulated. Heat transfer empirical correlations used and other details can be found in Appendix (A).
\[ Q_{\text{loss}} = Q_{\text{conv},g} + Q_{\text{rad},g} = Q_{\text{cond}} = Q_{\text{conv},c} + Q_{\text{rad},c} \] (18)

After heat losses are calculated, flame temperatures are estimated from equilibrium calculations using the adiabatic assumption, using CHEMKIN-PRO (Reaction Design, Release 15131), from the output \( O_2 \) mole fraction \( (X_{O_2}) \). After adiabatic flame temperatures are estimated, heat-loss-corrected temperatures are calculated assuming a one-dimensional heat loss model as described in Figure 23 with the accompanying calculations to heat loss correction.

![Figure 23: Corrected temperature calculation procedure](image)

2.4.5. Nitrogen Oxides

The calculation of nitrogen oxides (\( \text{NO}_x \)) is done by summing the dry mole fractions of nitrogen oxide (NO) and nitrogen dioxide (\( \text{NO}_2 \)), and usually converting to parts per million (ppm). Afterwards, correction to 15% \( \text{O}_2 \) as a reference is made to remove ambiguity when comparing different sets of experimental data. Calculation of \( \text{NO}_x \) \( @15\% \text{ O}_2 \) is shown by
where $\chi_{NO_{x}, dry}|_{15\% O_2}$ is the molar fraction of nitrogen oxides, on dry basis, corrected to 15% O$_2$, in parts per million (ppm)

$\chi_{NO, dry}$ is the molar fraction of nitrogen oxide (NO) on dry basis, in ppm

$\chi_{NO_2, dry}$ is the molar fraction of nitrogen oxide (NO$_2$) on dry basis, in ppm

$\chi_{O_2, %, dry}$ is the molar fraction of oxygen (O$_2$) on dry basis, in percent

### 2.5. Velocity Field and Flame Structure Measurements

In order to characterize the velocity field in the combustor area and calculate different turbulence parameters, particle image velocimetry (PIV) was utilized. In addition, to characterize the flame shape, define the premixed flame regime and calculate the flame zone volume for later uses in chemical kinetics modeling (Chapter 3), planar laser-induced fluorescence of OH radicals (OH-PLIF) was simultaneously measured. A schematic of the simultaneous PIV / OH-PLIF measurement alignment is shown in Figure 24.
2.5.1. Particle Image Velocimetry (PIV)

The basic principle of PIV is the calculation of fluid velocities by measuring the velocity of tracing seeded particles through measured displacements and predetermined time intervals. Opposed to laser Doppler velocimetry (LDV), one-camera PIV is capable of instantaneous two-dimensional mapping of two-component fluid velocities across a specified measurement (target) plane, which makes it a whole-flow-field measurement technique.

As shown in Figure 25, double-pulsed Nd:YAG 532 nm laser is produced from a PIV laser head (New Wave Research Solo 120, up to 120mJ/pulse) through proper light sheet optics to give a laser sheet (480 μm thickness FWHM) that illuminates a target plane (0.75” × 2.48”)

Figure 24: Simultaneous PIV and OH-PLIF measurement
(Figure 26) inside the flow section through the optical access fused-silica windows. After mixing with the methane/air pre-mixture, brown aluminum oxide powder (AOB500G, 500-Mesh ~ 25 µm) particles are seeded with the flow at the bottom of the combustor controlled and fine-tuned by a flow control valve. The seeded powder particles were initially prepared, dispersed dry and continuously fed by a fluidized bed aerosol generator (TSI 9310), with bronze de-agglomerating beads (grade 60-P), and a continuous cloud of particles was formed.

![Figure 25: PIV measurement principle](http://www.dantecdynamics.com/measurement-principles-of-piv)
When the seeded particles are illuminated by the laser sheet, scattered light from the two laser exposures is captured by the laser CCD camera (LaVision Imager Intense, 12 bit, 1376 × 1040 pixels, double-frame, double-exposure) into two different frames. Both the laser head and CCD camera were triggered at 2 fps using a PIV-synchronizer, which was in turn triggered by a 3.1 MHz arbitrary waveform generator (Stanford Research Systems). The camera was fitted with a narrow bandpass interference filter (centered at 532 nm with 10 nm FWHM, transmission = 90%) to reject flame emission. A camera shutter (LaVision) was used to limit intensity from the second frame.

2.5.2. Velocity Field Post-processing

To post-process the PIV measurements, DaVis (version 8) software uses statistical correlation techniques in small sub-area divisions of the target plane, referred to as the
interrogation windows (32 × 32, with an overlap of 50%), between the two frames to indicate the
displacements of seeding particles. Knowing the time separation between the two laser flashes
(10 μs), velocity of seeding particles across the target plane can be calculated through the simple
relation:

\[ V = \frac{\Delta X}{\Delta t} \]  

(20)

where \( V \) is the velocity of seeding particle,

\( \Delta X \) is the displacement of seeding particles between the two frames, shown in Figure 25

\( \Delta t \) is the time interval between the two frames, shown in Figure 25

A challenge that faces proper PIV measurement is providing the appropriate size of
seeding particles. Small size allows good tracing of seeding particles to the flow and more
accurate representation of the flow velocities. However, small seeding particles scatter less laser
intensity, hence lower accuracy of camera imaging. As the particle size goes up, intensity of the
scattered laser increases, but the momentum of the particles affect its representation of the flow
as inertial forces cause the particles to deviate from the flow at sudden changes in direction and
velocity at different points. Therefore, proper sizing of the particles have been done.

Another challenge is involved with the quantity of seeding particles. As the quantity of
seeding particles increases, it covers the area in higher densities. However when quantity
increases excessively the particles become more unrecognizable when correlations are executed
by the PIV processing software as the packing density is too high. Another disadvantage with
high particle density in combustion lies within the fact that as seeding particles are heated by the
combustion flames, they tend to stick on the laser access windows. When they stick on the
window surfaces, light scattering at those particles causes much noise and affects the camera
imaging. This also puts a limitation on the PIV measurement time, as adhesiveness of particles is proportional to the overall time of running seeding particles through the combustor.

Using PIV, velocity field was measured through the target plane. A validation rate, defined by ratio of the interrogation windows resided by velocity vectors to empty ones, was reported to be more than 95% for all velocity measurements. Turbulence parameters can then be calculated from the velocity by using the equations below.

\[
\begin{align*}
    u(t) &= \bar{U} + u'(t), \quad v(t) = \bar{V} + v'(t) \\
    \bar{U} &= \frac{1}{N} \sum_{i}^N u(t), \quad \bar{U} = \frac{1}{N} \sum_{i}^N u(t) \\
    u'(t) &= u(t) - \bar{U}, \quad v'(t) = v(t) - \bar{V} \\
    u_{rms} &= \sqrt{\frac{1}{N} \sum_{i}^N (u'(t))^2}, \quad v_{rms} = \sqrt{\frac{1}{N} \sum_{i}^N (v'(t))^2} \\
    V_t' &= \sqrt{(v_{rms}^2 + 2u_{rms}^2)/3}, \quad \bar{V}_t \approx \sqrt{(2\bar{U}^2 + \bar{V}^2)} \\
    TI &= \frac{V_t'}{\bar{V}_t} \\
    R_{vx}(x, y, r) &= \frac{v'(x, y, t) \cdot v'(x + r, y, t)}{v_{rms}(x, y) \cdot v_{rms}(x + r, y)} \\
    \ell_{t,vx}(x, y) &= \int_{0}^{\infty} R_{vx}(x, y, r)dr \\
    \tau_{t,vx} &= \ell_{t,vx}/V_t' \\
    R_{vy}(x, y, r) &= \frac{v'(x, y, t) \cdot v'(x, y + r, t)}{v_{rms}(x, y) \cdot v_{rms}(x, y + r)} \\
    \ell_{t,vy}(x, y) &= \int_{0}^{\infty} R_{vy}(x, y, r)dr \\
    \tau_{t,vy} &= \ell_{t,vy}/V_t'
\end{align*}
\]
As in equation (21), each flow turbulent velocity component can be presented by Reynolds averaging as the sum of a mean velocity (U) and a velocity fluctuation (u’). The mean velocity (U) can be calculated by averaging the velocities at a point through the selected time interval, as seen in equation (22). Velocity fluctuations are then the difference between turbulent velocities and their mean values; equation (23). The root-mean-square (RMS) velocity fluctuation in each direction is then defined as the square root of the average squares of the velocity fluctuations at each instant, as in equation (24). The absolute turbulence intensity is defined as the square root of the average of the three RMS components. Since the jets are statistically axisymmetric, the circumferential mean velocity vanishes and the RMS circumferential velocity component (w_{rms}) is assumed to be equal to the radial (transverse) velocity component (v_{rms}), hence equation (25), based on 3-D averaging; also previously used in a similar study by [97]. It is worth mentioning that another set of calculations based on 2-D averaging was compared and no significant difference in results was reported. The relative turbulence intensity (or shortly turbulence intensity, TI) is equal to the ratio of absolute turbulence intensity to the mean velocity; equation (26).

To establish a successful chemical kinetics model to study the turbulence-chemistry interactions and estimate the NOx emissions, performed by CHEMKIN-PRO and explained later in the next chapter, a mixing time scale (eddy turnover time scale) should be calculated from the turbulence parameters. This eddy turnover time scale can be defined by the ratio of a mixing length scale divided by the absolute turbulence intensity, as shown in equation (29). This length scale can be calculated by obtaining two-point velocity correlations between different points in
the target plane, as in equation (27). Inside the same eddy, two points are correlated and the correlation parameter is larger than zero. As the distance between the two points increases, the correlation parameter decreases until it reaches zero. As illustrated in Figure 27, the integral length scale can be calculated using the area under the curve until the correlation parameter crosses a value of zero, as calculated by equation (28). Since the flow is primarily characterized by axial velocities, the length scales and the corresponding time scales are calculated using axial velocities and correlating them both in the transverse direction (equations (27) - (29)) and in the longitudinal direction (equations (30) - (32)). Probability distributions can be generated from the time scales as will be shown later with accompanying calculation.

![integral length scale calculation](image)

**Figure 27: Integral length calculation from correlation parameters**

### 2.5.3. Planar Laser-Induced Fluorescence of OH radicals (OH-PLIF)

To characterize the flame zone structure and dimensions, OH-PLIF was used, simultaneously with PIV measurement, and aligned as was shown in Figure 24. The OH-PLIF diagnostic system consists of an Nd:YAG laser (Spectra-Physics LAB-170-10), used to pump a tunable dye laser (Sirah Cobra-Stretch) at 532 nm with energy of about 450 mJ/pulse. The output
of the tunable pulsed dye laser was frequency-doubled to give about 15 mJ/pulse at 283.92 nm with a pulse duration of approximately 10 ns. A spherical-cylindrical lens combination was used to generate a beam sheet of 60 mm height and approximately 250 μm thickness, to cover the same target plane measured by PIV. The excitation of OH radicals involved tuning the laser beam to the Q2(8) and Q1(9) lines in the (1, 0) A2Σ+ - X2Π transition, while broadband detection of the vibrationally-relaxed (1, 1) and (0, 0) fluorescence signal near 310 nm was made using a longpass glass filter (WG-305) and a colored glass filter (UG11). Typically, a 50 mm x 50 mm field of view was employed using a UV-camera lens (UV-Nikkor, 105 mm, f/4.5) that focused the image onto an intensified CCD camera (Princeton Instruments PI-MAX 4). The camera was synchronized with the laser at 4 fps for the digital collection of fluorescence generated by the OH radicals, when they absorb laser energy, as shown in Figure 28.

![Figure 28: PLIF measurement principle](image)

When the OH-PLIF technique targets the OH radicals, the detected fluorescence signal is proportional to the number of OH radicals within the target plane. Hence when the premixed

---

methane/air mixture is burnt, OH radicals concentration increases as a result of the combustion reaction. Hence OH radical concentration are very low (mainly zero) before the flame front, where gases are still unburnt, and increase suddenly at the flame front to a much larger value at the post-flame zone, so the flame fronts could be distinguished and characterized from OH radical mapping obtained from OH-PLIF measurements, for example the flame shown in Figure 29. From such flame shape, the flame regime can be distinguished, to better analyze the interaction between turbulence and combustion chemistry. Also flame zone volumes can be calculated, for calculation of flame zone residence times, a parameter definitely useful in chemical kinetics modeling, as will be seen in the next chapter.

![Figure 29: Extraction of flame front shape from an OH-PLIF instantaneous image](image)

In the next chapter, the chemical kinetics modeling approach followed will be discussed and explained. Later, results obtained both experimentally and numerically will be compared and validated, and then an analysis of the effect of turbulence is made and conclusions will be withdrawn.
Chapter 3

Chemical Kinetics Modeling

3.1. Introduction

In many combustion processes, formation and destruction of pollutant combustion products are controlled by rates of chemical reactions. Chemical kinetics is the branch of physical chemistry that specializes in studying the elementary reactions and their rates, and is needed for understanding the underlying chemical processes in detail. In the past few decades, chemical pathways and mechanisms have been developed to keep track of the detailed chemical kinetics to the best approximation possible.

In combustion, of the most important to keep track of is nitrogen oxides (NO\textsubscript{x}). However, since the reactions forming the intermediate species leading to NO\textsubscript{x} formation are slow, the use of chemical equilibrium approximations are useless, since the residence times in typical combustors (a few milliseconds) are insufficient for NO\textsubscript{x} to reach chemical equilibrium (in order of seconds). Chemical kinetics is used instead to study NO\textsubscript{x} formation, by the calculation of elementary reaction rates and mechanisms.

3.2. Chemical Kinetics Schemes

Many reduced and full kinetics schemes (lists of elementary chemical reactions and associated rate constant expressions) have been developed to model the formation of NO\textsubscript{x} using
methane fuel. GRI-Mech is one of the most important optimized schemes used to track the reactions forming NOₓ and their rates, in methane and natural gas flames. Development of extensions of GRI-Mech is ongoing, to include more chemistry details and updated values of elementary reaction rates. The latest release of this scheme is GRI-Mech 3.0. It compiles 325 elementary chemical reactions and associated rate coefficient expressions and thermochemical parameters for the 53 species involved in them. It has been optimized for methane and natural gas fuel, and is used in the present study to model NOₓ with the current study conditions. The construction of chemical schemes, including GRI-Mech 3.0, needs many empirical studies of different rates of chemical reactions, gathered from several studies and universities and institutions have been collaborating for the development of such mechanisms and schemes.

However, the application of GRI-Mech needs many numerical efforts and very lengthy computations. Numerical packages have been developed and dedicated to the numerical computation details of chemical kinetics, such as CHEMKIN, Cantera, and Chemical Workbench. CHEMKIN software, originally developed by Sandia National Laboratories and now developed at Reaction Design, has been one of the most reliable software used for research and industry, and has been proven to model NOₓ to a good approximation with improving agreement with experimental results. CHEMKIN-PRO (Release 15131 - July 27, 2013), an upgrade to CHEMKIN, is used for modeling NOₓ in the present study and analyzing intermediates and conditions to explain the effect of turbulence on NOₓ in lean premixed combustion flames.

3.3. Chemical Kinetics Modeling

To model chemical kinetics involved in formation and destruction of molecules, atoms
and radicals, chemical kinetics coupled with fundamental conservation principles were used to
develop many reactor models. A few examples of chemical reactor models used are shown in
Figure 30. The choice of reactor model depends on the nature of combustion and the state of
mixedness inside. For steady-state steady-flow open systems, the most two important widely
used reactors are perfectly-stirred (or well-stirred) reactor (PSR) and plug-flow reactor (PFR).
Many complex flows are usually modeled using these reactors as building blocks for a large
chemical reactor network. The accuracy of models and their degree of representation of actual
physical chemistry depends on the number of reactors, their distribution and inputs to each.

Figure 30: Simple chemically reacting systems; (a) constant-pressure, fixed mass; (b) constant-volume, fixed
mass; (c) well-stirred reactor; (d) plug-flow reactor. [98]
3.3.1. Perfectly-Stirred Reactor (PSR) Model

A PSR (Figure 30c) is an ideal reactor assumes a state of perfect mixing and homogeneous composition inside the control volume, and is therefore classified as a zero-dimensional (0-D) reactor. Figure 31 illustrates a PSR control volume with the parameters involved.

Figure 31: A perfectly-stirred reactor (PSR) Model

The main governing equations for a PSR are the mass conservation equations for arbitrary species $i$, for the integral control volume shown in Figure 31, and are represented by equation (33):

$$\frac{dm_{i,cv}}{dt} = \dot{m}_i'V + \dot{m}_{i,in} - \dot{m}_{i,out}$$  \hspace{1cm} (33)

Rate at which mass of $i$ accumulates  \hspace{1cm} Rate at which mass of $i$ is \hspace{1cm} Mass flow of $i$ into \hspace{1cm} Mass flow of $i$ out of
For steady-state operation, the previous equation becomes:

$$\dot{ω}_i MW_i V + \dot{m}(Y_{i,in} - Y_{i,out}) = 0 \quad \text{for } i = 1, 2, \ldots, N \text{ species}$$ (34)

Assuming the outlet mass fractions to be equal to the mass reactors inside the reactor since the composition inside the reactor is homogenous, the species production rates ($\dot{ω}_i$) are functions of temperature $T$ and molar fractions $X_i$, and not time $t$. Therefore, along with the energy equation (equation (35)), a set of nonlinear algebraic equations are formed and could be solved simultaneously to calculate the outlet species concentrations and temperatures.

$$Q = \dot{m}(h_{out} - h_{in})$$ (35)

It is also worth mentioning that a mean residence time ($t_R$) is commonly defined by equation (36), with calculation of the mixture density from the ideal gas law, equation (37).

$$t_R = \frac{\rho V}{\dot{m}}$$ (36)

$$\rho = PMW_{mix}/R_u T$$ (37)

As observed from the previous equations, composition and temperature inside a PSR depends on $t_R$. As $t_R$ decreases to a certain limit, at which chemical time scales become comparable, combustion can not be sustained and a flame blows out. This limit is known as the blowout limit and is important for steady reliable operation of gas turbine combustors.

Inside the flame zone, since combustion occurs rapidly at the flame front, perfect mixing can be assumed and a PSR is used to model chemical kinetics inside. Combustion in the post-
flame zone is however a time/distance dependent process and is hence modeled by a PFR as shown later. High-velocity jet combustion also use PSR’s to study flame stabilization, since a PSR assumes perfect mixing between reactors and following intermediates and products.

3.3.2. Plug-Flow Reactor (PFR) Model

On the other hand, a PFR (Figure 30d) is an ideal reactor that totally ignores mixing and diffusion in the flow (axial) direction, but assumes perfect mixing and uniform properties in the transverse (radial) direction. This means that any cross section can be represented by a single point completely characterized by a single velocity, temperature and composition. Using conservation equations across an infinitesimal distance (Δx), as shown in Figure 32, a set of ordinary differential equations (equations (38), (39) and (40)) can be formulated and integrated using an appropriate set of initial conditions.

\[
\frac{d\rho}{dx} = \left(1 - \frac{R_u}{c_p MW_{mix}}\right) \rho^2 \left(\frac{1}{A} \frac{dA}{dx}\right) + \frac{\rho R_u}{v_x c_p MW_{mix}} \sum_{i=1}^{N} \omega_i MW_i \left(\frac{h_i}{MW_i} - \frac{MW_{mix}}{MW_i} c_p T\right) \\
\frac{P}{P_{mix}} \left(1 + \frac{v_x^2}{c_p T}\right) - \rho v_x^2
\]

\[\text{(38)}\]

\[
\frac{dT}{dx} = \frac{v_x^2}{\rho c_p} \frac{d\rho}{dx} + \frac{v_x^2}{c_p} \left(\frac{1}{A} \frac{dA}{dx}\right) - \frac{1}{v_x \rho c_p} \sum_{i=1}^{N} \omega_i MW_i
\]

\[\text{(39)}\]

\[
\frac{dY_i}{dx} = \frac{\omega_i MW_i}{\rho v_x}
\]

\[\text{(40)}\]
Figure 32: Control volumes showing fluxes of mass, x-momentum, energy, and species for a plug-flow reactor (PFR). From [98]

The residence time \( t_R \) can be defined as

\[
\frac{dt_R}{dx} = \frac{1}{v_x} \tag{41}
\]

Since a PFR takes into consideration time accumulation of combustion products, it is classified as a one-directional (1-D) reactor and is suitable for estimation of the buildup of time-dependent species (such as NO\(_x\)) in the post-flame zone.
To model combustion without considering the transverse variation, and hence approximating the flame to a one-dimensional combustion process, the flame is divided into two zones. The first zone is referred to as the flame zone, and includes a volume in which the combustion region is well-mixed, around the flame front. This zone can be modeled as a PSR, since it assumes perfect mixing. After the flame front, the second zone is referred to as the post-flame zone, and can be modeled as a PFR, since the mixing is slower and NO\textsubscript{x} is slowly accumulated. The simplest form of a reactor network is then PSR + PFR, as shown in Figure 33. Several literature used a PSR + PFR reactor network to model lean premixed combustion, like work done by Elkady et al. [82].

![Chemical reactor network (PSR+PFR) used in [82]](image)

**Figure 33: Chemical reactor network (PSR+PFR) used in [82]**

### 3.3.3. Partially-Stirred Reactor (PaSR) Model

As shown above, a PSR assumes perfect mixing and hence infinite stirring frequency (turbulence level). In reality, mixing inside a combustor (also referred to as in-combustor stirring) is finite. Because in these conditions, the turbulent mixing rate is not fast compared to chemical kinetics, the degree of mixing can have a large impact on the reactor characteristics. To model finite mixing and effect of turbulence on combustion and associated chemistry interactions, the PSR should be improved to consider imperfect mixing. The imperfect-mixing (yet much more complicated) version is the partially-stirred reactor (PaSR).
To allow the relaxation of the fast turbulent mixing assumption implied in a PSR, the mean thermo-chemical properties inside a PaSR model are assumed to be variable at the molecular level, but however spatially homogeneous on the macro-scales (and hence, like a PSR, a PaSR is considered a 0-D model). The mixing process in a PaSR is characterized by the mixing frequency ($\omega$), which is the reciprocal of the turbulence time scale. The mixing frequency (or the corresponding mixing time scale) must be specified as an input to the PaSR, in addition to the mean reactor residence time which is the only input to a PSR.

The composition and temperature in a PaSR are described by a probability density function (PDF). A joint PDF in a turbulent reactive flow describes the probability of both progression variable $c$ (that represents both species concentration and temperature) and turbulent velocities [99-102]. The composition PDF in a PaSR is a subset of this joint velocity-composition PDF, since the flow field in the PaSR is assumed to be spatially homogeneous. Velocity fluctuations are also ignored, and the PDF is over scalars only, but it is not a delta function in scalar space because reactants, intermediates and products are not mixed at the molecular level.

Through the variation of level of turbulent mixing, a PaSR is bounded by other reactor models. A PaSR is bounded by the PSR model, in the special case of perfect mixing (zero mixing time) as the properties are homogeneously mixed at the molecular level. In this case, the joint scalar PDF degenerates to a delta function in the composition space, and then the only controlling time scale is the mean reactor residence time. On the other hand, when the mixing time becomes very large, mixing between gas pockets in the PaSR is negligible, and the PaSR consists of segregated reactive mixtures. In this case, the PaSR acts like a PFR, for no mixing
and relatively large mean residence times.

The mixing time scale in a PaSR \( t_{mix} \) allows fluid dynamics to control the molecular mixing and hence the chemical reactions, and is considered to be proportional to the turbulent eddy turnover time, as defined by the ratio between the turbulent kinetic energy to its dissipation rate [100], as

\[
\tau_{mix} = \frac{k}{\varepsilon}
\]

Since this represents the time scale of the energy-containing eddies, it can also be defined as:

\[
\tau_{mix} = \frac{\ell_t}{V_t}
\]

3.3.3.1. The Joint PDF Transport Equation

The chemical closure for the nonlinear governing equations in turbulent flow can be provided by considering the joint probability density function (PDF) of the flow variables. The joint PDF of scalars, \( P_\phi(\psi; \mathbf{x}, t) \), provides a complete statistical description of the chemical and thermodynamic state. For reactive flows involving many species, the dimensions of the PDF \( P_\phi(\psi_1, \psi_2, ..., \psi_\sigma; \mathbf{x}, t) \) are large, and then finite-difference solutions of the PDF transport equations are not practical because \( \sigma \) is large. Instead, a Monte Carlo algorithm was developed by Pope [101] to allow practical solutions of the PDF transport equation for turbulent reactive flows. The dependent variable in this stochastic simulation is represented by an ensemble of \( N \) particles, denoted by \( \phi^{(1)}, \phi^{(2)}, ..., \phi^{(n)}, ..., \phi^{(N)} \). Each particle is given a unique index, \( 1 \leq n \leq N \), but however no ordering is implied. The ensemble average of any function \( Q(\phi) \) is defined as
\[
\langle Q(\phi) \rangle \equiv \frac{1}{N} \sum_{n=1}^{N} Q(\phi^{(n)})
\]

(44)

It was shown [101] that for large \( N \), the ensemble average \( \langle Q(\phi) \rangle \) converges to the corresponding density-weighted average,

\[
\langle Q(\phi) \rangle \rightarrow \bar{Q}(\phi) = \int Q(\psi)\bar{P}_\phi(\psi)d\psi
\]

(45)

By integrating the governing equation of the single-point joint scalar PDF over the reactor volume, the resulting PDF transport equation for the PaSR is

\[
\frac{\partial \bar{P}_\phi(\bar{\psi}, t)}{\partial t} = -\sum_{\alpha=1}^{\kappa_{tot}} \frac{\partial}{\partial \psi_\alpha} \left\{ S_\alpha(\bar{\psi})\bar{P}_\phi(\bar{\psi}, t) \right\}
\]

\[+ \frac{1}{t_R} \sum_{i=1}^{M} \left\{ \bar{P}_{\phi,i}(\bar{\psi}, t) - \bar{P}_\phi(\bar{\psi}, t) \right\}
\]

\[- \sum_{\alpha=1,\beta=1}^{\kappa_{tot}} \frac{\partial^2}{\partial \psi_\alpha \partial \psi_\beta} \left\{ (\epsilon_{\alpha\beta} | \bar{\phi} = \bar{\psi}) \bar{P}_\phi(\bar{\psi}, t) \right\}
\]

(46)

The first two terms on the right hand side of equation (46) represent the effects of chemical reaction and the through-flow on the joint PDF, respectively. The last term represents the effect of micro-scale mixing on the PDF, which requires the use of a mixing model. The mixing model mimics the finite rate mixing of particles in the stochastic simulations. Two mixing models are widely used and considered:

1. Modified Curl’s Mixing Model
\[
\sum_{\alpha=1, \beta=1}^{k_{tot}} \frac{\partial^2}{\partial \psi_\alpha \partial \psi_\beta} \{ (\varepsilon_{\alpha\beta} | \bar{\Phi} = \bar{\Psi}) \bar{P}_\Phi(\bar{\Psi}, t) \}
\]

\[
= \frac{1}{\tau_{mix}} \left\{ \int_{\psi', \psi''} \bar{P}_\Phi(\psi', t) \bar{P}_\Phi(\psi'', t) H(\psi', \psi'' | \bar{\Psi}) d\psi' d\psi'' \right. 
- \bar{P}_\Phi(\bar{\Psi}, t) \right\}
\]

where \( H \) is the transitional probability defined as \( H(\psi', \psi'' | \bar{\Psi}) = \{1/|\psi'' - \psi'|\} \) for \( \psi \in [\psi', \psi''] \) otherwise 0.

2. Interaction-by-Exchange-with-the-Mean (IEM) or the Linear-Mean-Square-Estimation (LMSE) Model

\[
\sum_{\alpha=1, \beta=1}^{k_{tot}} \frac{\partial^2}{\partial \psi_\alpha \partial \psi_\beta} \{ (\varepsilon_{\alpha\beta} | \bar{\Phi} = \bar{\Psi}) \bar{P}_\Phi(\bar{\Psi}, t) \}
\]

\[
= \frac{C_\Phi}{2\tau_{mix}} \frac{\partial}{\partial \psi_\alpha} \{ (\bar{\Psi} - \Phi) \bar{P}_\Phi(\bar{\Psi}, t) \}
\]

where \( C_\Phi \) is a constant parameter for the model.

### 3.3.3.2. PaSR Reactor Equations

Similar to the PSR, the PaSR consists of an adiabatic chamber having \( M \) inlet streams and one outlet. Steady flows of reactants are introduced through the inlets with given gas compositions and temperatures. In order to represent the evolution of the PDF properly by a stochastic scheme, PaSR addresses all problems in a transient manner. The average properties of
the PaSR are obtained from the ensemble of particles inside the reactor. Each particle is regarded as an independent PSR and interacts with others only through the molecular mixing process. Therefore, the conservation of energy and species is applied to an individual particle rather than to the reactor. Hence the conservation equations for a particle are similar to that of a PSR:

\[ \frac{d\langle M \rangle}{dt} = \frac{d((\rho)V)}{dt} = \sum_{i=1}^{M} \dot{m}_i - \dot{m}_o = 0 \]  

(49)

\[ \frac{dY_k^{(n)}}{dt} = \frac{1}{\dot{m}_o t_R} \sum_{i=1}^{M} \{ \dot{m}_i (Y_{i,k} - \langle Y_k \rangle) \} + \frac{W_k \omega_k^{(n)}}{\rho^{(n)}} \]  

(50)

\[ \frac{dT^{(n)}}{dt} = \frac{1}{C_p^{(n)} \dot{m}_o t_R} \sum_{i=1}^{M} m_i \left( \sum_{k=1}^{k_g} Y_{i,k} (h_{i,k} - \langle h_k \rangle) \right) - \frac{\sum_{k=1}^{k_g} W_k \omega_k^{(n)} h_k^{(n)}}{\rho^{(n)} C_p^{(n)}} \]  

(51)

Again, the mean residence time can be defined by equation (36).

3.3.3.3. Stochastic Simulation

To solve for properties in a PaSR, a time marching scheme with a time-step size of \( \Delta t \) is used, and the stochastic simulation is carried out by the following sequential procedures with \( N \) statistical particles:

1. Through-flow (Convection)
2. Molecular Mixing

First, the number of correct particles in a time step is chosen as \( N_c = N \times \frac{\Delta t}{t_R} \). The properties of these particles are set from the stochastic ensemble as the properties of the inlet mixture. Then, using the modified Curl’s mixing model, a number of particles are chosen to mix
with each other. The number of particles is determined by: \[ N_m = C_m N \times \frac{\Delta t}{\tau_{m}} \], where \( C_m \) is parameter for the modified Curl’s model. The next step is the computation of chemical kinetics for each particle by integrating the species and energy equations (equations (50), (51)) over a period of \( \Delta t \). The same procedures are repeated for each time step until the end of the simulation time is reached.

In the present study, a modified Curl’s model with a scaling factor of 2, as recommended by a Reaction Design application note [103] including a similar case of a premixed methane/air combustor. A time step of 0.1 ms is used with a total simulation time of 30 ms, and 500 statistical particles.

### 3.3.4. Modeling the Flame

To model the flame in a one-dimensional approximation, the flame zone is modeled by a PaSR instead of the commonly-used PSR to consider turbulence effects. Therefore the reactor network is PaSR + PFR, as shown in Figure 34. A snapshot of the diagram window of CHEMKIN-PRO showing the reactor network is also shown in Figure 35. Mixing times calculated from PIV measurement, as previously explained in chapter 2, were used for modeling. Residence times were calculated from equation (36). The volumes of flame and post-flame zones (that appear in equation (36) were calculated by approximating the cross section for each of the two zones as a square, whose side equals to the spacing between nozzle holes (= 0.25”), and using a flame height, calculated from average PLIF imaging, as shown before in chapter 2.
To further investigate the effect of turbulence, a parametric study was performed using a range of mixing times (0.01 ms, 0.02 ms, 0.05 ms, 0.1 ms, 0.2 ms, 0.5 ms) was investigated, and flame zone residence time was also varied (2.632 ms, 3.948 ms and 5.264 ms - corresponding to flame heights of 0.6”, 0.9” and 1.2”, respectively).

Later in chapter 4, the modeling results are compared to the experimental findings. The effect of turbulence is analyzed using parametric studies of mixing time scales and the effect of their variation on temperatures and oxygen atom concentrations.
Chapter 4

Results and Discussion

In this chapter, results from both experimental and modeling approaches are presented and compared to similar cases from literature. The modeling results are then compared to the experimental ones. Different methods for correction are also presented. Results are then analyzed and further discussion is made.

4.1. Experimental Results

Using the experimental approach described in chapter 2, data regarding combustion emissions, PIV results, OH-PLIF measurements are extracted and accompanying calculations are performed.

4.1.1. NO<sub>x</sub> Emissions

For both nozzles (48N0110 and 48T0110), nitrogen oxides and other emissions are measured and correction of NO<sub>x</sub> to 15% O<sub>2</sub> (equation (19)) is made, for a range of equivalence ratios (about 0.54 to 0.85). Adjusting equivalence ratios to O<sub>2</sub> emissions (as previously shown in chapter 2), was applied, and nitrogen oxides emissions are shown versus O<sub>2</sub>-adjusted equivalence ratios in Figure 36. Using O<sub>2</sub>-adjusted data is considered a basis for comparison, as shown for example in the work of Leonard and Correa [81] and Elkady et al. [82].
Using CHEMKIN-PRO (release 15131), flame temperatures were calculated, using chemical equilibrium assumptions, for both nozzles over the range of equivalence ratios studied. Heat loss correction calculations were made to correct flame temperatures and are shown briefly in Figure 23 and with extensive details in Appendix A. Disregarding heat loss correction in estimating flame temperatures may lead to artifacts including lower NO\textsubscript{x} for the adiabatic temperature calculations as can be seen in comparison to the heat-loss-corrected measurements.

Adjusting to O\textsubscript{2} emissions (Figure 23) is applied to estimated flame temperatures. NO\textsubscript{x} emissions are plotted versus O\textsubscript{2}-adjusted flame temperatures, in Figure 37 and Figure 38 with and without heat loss correction.
Figure 37: NO\textsubscript{x} vs. O\textsubscript{2}-adjusted flame temperatures for the two nozzles (adiabatic)

Figure 38: NO\textsubscript{x} vs. O\textsubscript{2}-adjusted flame temperatures for the two nozzles (with heat loss correction)
The O₂-adjusted equivalence ratios and flame temperatures are the quantities used for comparison and validation with different literature, as also used in previous similar work (e.g. [81, 82]), to relate different sets of data at different oxygen levels.

As seen in Figure 38, there is almost no change in the NOₓ levels due to turbulence level increase. The slight difference in results may be due to experimental errors. The two sets of data can even be presented by one curve fit, as shown in Figure 39.

Leonard and Correa [81] used a configuration similar to the current experimental work in order to investigate the emissions from lean premixed methane flames in jets from perforated
plate with the same unburnt jet velocity (= 10.59 m/s). They performed their tests at several inlet reactant temperatures (100°F, 500°F, and 650°F). Results from inlet temperature of 500°F (=533K) was chosen for comparison with this study (~495 K). At this selected inlet temperature, they showed results at 10.4 atm. According to their observation, NOₓ molar fraction had a dependence on pressure (P) more in the form of \( \sqrt{P} \) as equivalence ratio increases. Therefore, to extrapolate their data from 10.4 atm to 5 atm (present study combustor pressure), a factor of \( \sqrt{10.4/5} \) was used to estimate their results at the same pressure of the current study of 5 atm, to at least observe comparison at higher equivalence ratios. However, measurements in their work were taken at 10 ms mean residence time, while the mean residence time here is around 24 ms, which causes significant differences in NOₓ emissions. As will be shown later, the chemical kinetics model agrees well with the NOₓ emissions measured in the present study (at mean residence time of 24 ms; Figure 72), and Leonard and Correa’s work (at mean residence time of 10 ms; Figure 73). The latter is taken as validation data for the current NOₓ results. Since NOₓ emissions changes with total residence time in a linear trend (Figure 77 to Figure 79), the ratio between modeling data at mean residence times of 10 ms and 24 ms, respectively, is used to extrapolate Leonard and Correa’s data to the present study mean residence times, and an agreement was observed, as in Figure 40.
Figure 40: NOₓ vs. O₂-adjusted Leonard and Correa’s data [81] (extrapolated to combustor pressure of 5 atm and mean residence time of 24 ms)

4.1.2. Flame Structure Characterization

After simultaneous PIV and OH-PLIF measurements were performed at three equivalence ratios (0.6, 0.66 and 0.78), instantaneous and average PLIF images can be used to characterize the flame shape. The flame front can be identified by the steep gradient in OH fluorescence signal intensities from very low values (dark) for unburnt reactants mixture to high values (bright) for burnt products. Examples of the instantaneous PLIF images for different conditions are shown in Figure 41 to Figure 46 for the 48N0110 and 48T0110 nozzles, respectively.
Figure 41: Examples of instantaneous PLIF for 48N0110, $\Phi = 0.60$

Figure 42: Examples of instantaneous PLIF for 48N0110, $\Phi = 0.66$
Figure 43: Examples of instantaneous PLIF for 48N0110, $\Phi = 0.78$

Figure 44: Examples of instantaneous PLIF for 48T0110, $\Phi = 0.60$
Figure 45: Examples of instantaneous PLIF for 48T0110, $\Phi = 0.66$

Figure 46: Examples of instantaneous PLIF for 48T0110, $\Phi = 0.78$
It can be seen from the previous figures that for higher equivalence ratios (0.78), combustion is coupled with acoustic instabilities, as also noticed as a fixed-tone noise during the operation of combustor. Acoustic instabilities cause the flame height to fluctuate over, and increase turbulence inside the flame in addition to flow turbulence and flame-generated turbulence, as will also be shown later from average turbulence levels. This effect is more pronounced in the 48T0110 nozzle than the 48N0110 one.

As shown in Figure 41 to Figure 46, because of relatively high turbulence intensities, the laminar flame front is highly corrugated. When the OH intensities at all points inside the measurement target plane are averaged over a specified period of time (around 200 images or 50 seconds), the turbulent average flame is characterized by a flame brush. This flame brush appears as a region between the reactants (dark region) and products (bright region) which is characterized by mid-intensities (grey region). The averaged OH-PLIF intensities are shown in Figure 47 and Figure 48 for the 48N0110 and 48T0110 nozzles respectively. The averaged flame can then be shown to have a minimum height ($h_{min}$) and a maximum height ($h_{max}$), which are the flame brush boundaries. It can be seen that the flame brush is not as clear at high equivalence ratios due to the fluctuation of the flame due to combustion instabilities as at low equivalence ratios. As shown in Figure 49, the flame zone volume is approximated as the volume of a rectangular parallelepiped whose base is a square, each side of which is equal to the spacing between holes ($s = 0.25" = 0.635$ cm), and its height is the average height of the flame = $\frac{1}{2} (h_{min} + h_{max})$. This volume is used to calculate the flame zone residence time which is a necessary input to the PaSR model for chemical kinetics modeling, and to its corresponding PSR model (an asymptotic limit check). Flame zone residence times for different conditions for both nozzles are listed in Table 1.
Figure 47: Calculation of flame height (48N0110) – (a) $\Phi = 0.60$, (b) $\Phi = 0.66$, (c) $\Phi = 0.78$

Figure 48: Calculation of flame height (48T0110) – (a) $\Phi = 0.60$, (b) $\Phi = 0.66$, (c) $\Phi = 0.78$
Figure 49: Rectangular parallelepiped flame zone

Table 1: Flame zone residence times

<table>
<thead>
<tr>
<th>$t_R$ (ms)</th>
<th>$\Phi = 0.6$</th>
<th>$\Phi = 0.66$</th>
<th>$\Phi = 0.78$</th>
</tr>
</thead>
<tbody>
<tr>
<td>48N0110</td>
<td>4.408</td>
<td>4.174</td>
<td>3.954</td>
</tr>
<tr>
<td>48T0110</td>
<td>3.634</td>
<td>2.966</td>
<td>2.856</td>
</tr>
</tbody>
</table>

From Figure 47 and Figure 48, it is observed that for each nozzle, as the equivalence ratio increases, the flame height decreases, and the flame brush thickness increases due to combustion instabilities and flame generated turbulence. This is due to the increase of turbulence intensities with equivalence ratios, as will be seen later in Table 2. If both nozzles are compared at the same equivalence ratio, the 48T0110 nozzle delivers shorter flames than the less turbulent 48N0110 nozzle, due to higher turbulence intensities (about 2-3 times), as will be shown in Table 2.

4.1.3. Velocity Field Characterization

From PIV measurements, instantaneous flame velocity fields were measured at a rate of 2 Hz. Average PIV velocity vectors are drawn on top of OH-PLIF intensities as shown in Figure
and Figure 51. The reduction of PLIF intensities from right to left is due to the absorption of laser intensities by OH radicals inside the combustor.

Figure 50: PIV vectors and OH-PLIF Contours for 48N0110

Figure 51: PIV vectors and OH-PLIF Contours for 48T0110
From OH-PLIF contours and PIV velocity vectors shown above, recirculation zones can be observed close to the nozzle surface between nozzle holes. Areas on the nozzle surface between the holes act as bluff bodies that create these recirculation zones and help stabilize the flame. The height of these recirculation zones is directly proportional with the flame height. As equivalence ratio increases, flame height decreases and the volume of recirculation zones consequently decreases. Velocities increase at the flame fronts, due to sudden decrease of densities from unburnt reactant mixture to hot combustion products. Velocity profiles show higher velocities at the nozzle hole centerlines than between nozzle holes, and velocities are more uniform across the combustor cross section far downstream of the nozzle surface.

4.1.4. Calculation of Turbulence Parameters

As shown before in chapter 2 (equations (24) through (31)), different turbulence parameters could be calculated. RMS Velocity fluctuations, absolute and relative turbulence intensities (TI) were calculated and a probability density functions (pdf) for TI values are shown in Figure 52 to Figure 59 for cold (unburnt) flow and for different equivalence ratios for the two nozzles. The pdf for each condition was calculated by forming a histogram of the number of occurrences of different TI values into 20 bins, and dividing by the sum of total number of occurrences, so that the area under the curve is equal to unity. The points selected for forming the pdf are those inside the flame region (up to the flame height, calculated before from OH-PLIF average intensity images. The density-weighted averages of TI for both nozzles at different conditions are listed in Table 2). As can be seen in cold flow for the 48N0110 and 48T0110 nozzles, the density-weighted average of TI are approximately equal to 28.5% and 73.5%, respectively. As the equivalence ratio increases, density-weighted average TI values increase.
Figure 52: TI for 48N0110 – \( \Phi = 0.60 \)

Figure 53: TI for 48N0110 – \( \Phi = 0.78 \)

Figure 54: TI for 48N0110 – cold flow

Figure 55: TI for 48N0110 – \( \Phi = 0.66 \)
Figure 56: TI for 48T0110 – Φ = 0.60
Figure 57: TI for 48T0110 – Φ = 0.78
Figure 58: TI for 48T0110 – cold flow
Figure 59: TI for 48T0110 – Φ = 0.66
It can be seen from Table 2 that average flame zone turbulence intensities from the 48T0110 nozzle are 2-3 times those from the 48N0110 nozzle. As equivalence ratios increase, turbulence levels significantly increase especially closer to stoichiometric conditions. This is due to flame-generated turbulence contribution, as well as combustion instabilities that occur at higher equivalence ratios.

Length scales and corresponding eddy-turnover time scales (also called mixing time scales) were calculated from equations (27) to (29), based on the transverse length scales, and from equations (30) to (32), based on the longitudinal length scales. Since the turbulence is anisotropic, the length scale values based on transverse and longitudinal direction are different. This basically means the turbulent eddies do not have the same dimensions in each direction, and are more ellipsoidal than spherical. Hence the mixing time scales from both directions are different, and is taken here as the average between both values. Probability density functions of the mixing time scales are shown in Figure 60 to Figure 71 and density-weighted averages of mixing times are tabulated in Table 3. It can be seen that mixing time scales from the “turbulent” 48T0110 nozzle are about 35% to 60% of those from the “normal” 48N0110 nozzle.
Figure 60: $\tau_{t,vx}$ PDF for 48N0110 – $\Phi = 0.60$

Figure 61: $\tau_{t,vy}$ PDF for 48N0110 – $\Phi = 0.60$

Figure 62: $\tau_{t,vx}$ PDF for 48N0110 – $\Phi = 0.66$

Figure 63: $\tau_{t,vy}$ PDF for 48N0110 – $\Phi = 0.66$

Figure 64: $\tau_{t,vx}$ PDF for 48N0110 – $\Phi = 0.78$

Figure 65: $\tau_{t,vy}$ PDF for 48N0110 – $\Phi = 0.78$
Figure 66: $\tau_{t,\nu x}$ PDF for 48T0110 – $\Phi = 0.60$

Figure 67: $\tau_{t,\nu y}$ PDF for 48T0110 – $\Phi = 0.60$

Figure 68: $\tau_{t,\nu x}$ for 48T0110 – $\Phi = 0.66$

Figure 69: $\tau_{t,\nu y}$ PDF for 48T0110 – $\Phi = 0.66$

Figure 70: $\tau_{t,\nu x}$ PDF for 48T0110 – $\Phi = 0.78$

Figure 71: $\tau_{t,\nu y}$ PDF for 48T0110 – $\Phi = 0.78$
Table 3: Probability density-weighted averages of mixing time scales

<table>
<thead>
<tr>
<th>$t_{\text{mix}}$ (ms)</th>
<th>$\Phi = 0.6$</th>
<th>$\Phi = 0.66$</th>
<th>$\Phi = 0.78$</th>
</tr>
</thead>
<tbody>
<tr>
<td>48N0110</td>
<td>0.378</td>
<td>0.492</td>
<td>0.310</td>
</tr>
<tr>
<td>48T0110</td>
<td>0.149</td>
<td>0.170</td>
<td>0.204</td>
</tr>
</tbody>
</table>

4.2. Chemical Kinetics Modeling Results

After calculating the residence times and mixing times of the flame zone, CHEMKIN-PRO was used to predict NO$_x$ emissions from both nozzles, using chemical kinetics. As explained earlier in chapter 3, the flame zone was modeled by a partially-stirred reactor (PaSR) model, and the post-flame zone was modeled by a plug-flow reactor (PFR). As noted before, using a perfectly-stirred reactor (PSR) model for the flame zone would assume a perfect mixing and hence infinite turbulence intensity and zero mixing time. As mixing time inside the flame zone decreases, it reaches PSR as a limit for zero mixing times and this will be shown in the results below. On the other hand, using a plug-flow reactor (PFR) for the flame zone would assume no axial mixing and hence zero turbulence intensity and infinite mixing time. As mixing time increases, it would reach PFR as a limit. Hence both PSR and PFR should be bounding limits for the PaSR mixing time variation. This will be shown in the results below.

4.2.1. NO$_x$ Prediction

Emissions from CHEMKIN-PRO are calculated on wet basis, and hence were converted to a dry basis from equation (17), and NO$_x$ are calculated and corrected to 15% O$_2$ from equation (19) for comparison versus experimental results. NO$_x$ emissions corrected to 15% O$_2$ are shown for both nozzles versus flame temperatures in Figure 72. Modeling results in the previous figures
use a total (flame + post-flame) mean residence time of about 24 ms, which is the same from the present study configuration and conditions. Experimental results are also plotted on Figure 72 for comparison. Figure 72 show that the modeling results show good agreement with experimental for flame temperatures adjusted to O₂ emissions.

![Comparison of experimental and modeling results (NOₓ vs. O₂-adjusted flame temperatures)](image)

**Figure 72: Comparison of experimental and modeling results (NOₓ vs. O₂-adjusted flame temperatures)**

The same model was used to estimate NOₓ emission for a case reported by Leonard and Correa [81], where the residence time was 10 ms. A comparison of the results could be shown in Figure 73. As noticed, the modeling results show good agreement with Leonard and Correa’s work at the same conditions.
Figure 73: Modeling results at 10 ms mean residence time compared to Leonard and Correa’s results (extrapolated to 5 atm)

4.2.2. Analysis of Effect of Turbulence

Temperatures and NOx emissions, as predicted from the chemical kinetics model (PaSR + PFR), are plotted versus total residence times in Figure 74 to Figure 79, for the two nozzles for Φ = 0.60, 0.66 and 0.78.

As seen in Figure 74, flame temperatures are higher for the 48T0110 nozzle inside the flame zone (modeled by PaSR). The increase of flame zone temperature with increase of turbulence level can be explained by the fact that enhancing the turbulence level inside the flame zone increases the mixing between the premixed reactants and the hot combustion intermediates and products and hence increases the flame zone temperature. However, after the transition from the flame zone to the post-flame zone, temperatures quickly reach equilibrium values and hence
the temperatures in the post-flame zones are the same between the two nozzles.

It can be seen from Figure 77 to Figure 79, that NO\textsubscript{x} emissions are higher in the 48T0110 nozzle than the 48N0110 one. It can be seen from the figures that inside the flame zone, different NO\textsubscript{x} emissions are due to different NO\textsubscript{x} formation rates inside the flame zone. Because the contribution of the prompt NO route is only favored for rich combustion, it has negligible significance in lean premixed combustion, and the main contributions to NO formation are those of the thermal and N\textsubscript{2}O-intermediate routes.

Through the N\textsubscript{2}O-intermediate NO formation route, NO formation rate is directly proportional to oxygen atom molar fractions, as seen in equations (5) and (6). Formation rate of NO also increase exponentially with increase of flame zone temperature, due to the dependence of reaction rate constant on flame temperature, as shown below

\[ k(T) = A \exp(-E_A/R_u T). \]  

From the thermal NO formation route, it can concluded that rate of formation of NO can be approximated as

\[ \frac{d[NO]}{dt} = 2k_f [O]_{eq} [N_2]_{eq}, \]  

where \( k_f \) is the forward reaction rate constant of equation (1). It can be seen that in both N\textsubscript{2}O-intermediate and thermal NO routes, NO formation increases for higher temperatures and/or higher oxygen atom levels.

Since both flame zone temperatures (Figure 74, Figure 75 and Figure 76) and oxygen atom levels (Figure 80) are higher in the 48T0110 nozzle than the 48N0110 one, rate of formation of NO is higher, and hence NO\textsubscript{x} emissions are higher for the same flame zone.
residence time, as shown in Figure 77, Figure 78 and Figure 79. The relative difference is more significant for lower NOx emission levels (lower Φ). It becomes negligible at Φ = 0.78.

After the transition from the flame zone to the post-flame zone, temperature and oxygen atom levels quickly reach equilibrium values, and hence rate of formation of NO remains constant for the rest of the post-flame zone, as well as the same for both nozzles. This explains the linear trend of NO formation over total residence time variation, shown in Figure 77, Figure 78 and Figure 79. This linear trend allowed for the extrapolation of Leonard and Correa’s work [81] for validation with data from the present study.

Figure 74: Temperature vs. total residence time for the two nozzles at Φ = 0.60
Figure 75: Temperature vs. total residence time for the two nozzles at $\Phi = 0.66$

Figure 76: Temperature vs. total residence time for the two nozzles at $\Phi = 0.78$
Figure 77: NO\textsubscript{x} versus total residence time for the two nozzles at $\Phi = 0.60$

Figure 78: NO\textsubscript{x} versus total residence time for the two nozzles at $\Phi = 0.66$
Figure 79: NO\textsubscript{x} versus total residence time for the two nozzles at $\Phi = 0.78$

Figure 80: Oxygen atom levels inside the flame zone versus equivalence ratio for the two nozzles
4.2.3. Modeling Parametric Studies

From the previous figures, the effect of turbulence from modeling results can be noticed. The “turbulent” 48T0110 nozzle gives slightly more NO\textsubscript{x} emissions than the “normal” 48N0110 nozzle. Contributions to this effect can be due to changing mixing times, changing flame zone residence times (through changing flame height) and changing total residence times. To more understand each effect separately, a parametric study of only each of mixing time scales, flame zone residence times and total residence times is done.

4.2.3.1. Effect of Turbulent Mixing Time

4.2.3.1.1. Variation of PaSR Mixing Time

For a constant flame zone residence time of 3.948 ms (corresponding to a flame height of 0.9”), the effect of mixing time scale on temperatures and NO\textsubscript{x} emissions is shown in Figure 81 and Figure 82, respectively. Results at mixing times of 0.5 ms, 0.2 ms, 0.1 ms, 0.05 ms, 0.02 ms, 0.01 ms and 0 (PSR) were studied. As noticed from the figures, decreasing mixing time causes a slight increase in temperature, and relatively more significant increase in NO\textsubscript{x}. When the mixing time decreases from 0.1 ms to 0.01 ms, NO\textsubscript{x} increases by about 10% to 38%, depending on equivalence ratio. As mixing time decreases from 0.01 ms to the theoretical limit of a PSR (zero mixing time), the change in NO\textsubscript{x} emissions is negligible (0.2% to 4.3%). The same results are plotted again in Figure 83 and Figure 84 using equivalence ratios for the X-axis for convenience.
Figure 81: Temperatures at end of post-flame region vs. turbulent mixing time (flame zone residence time = 3.948 ms)

Figure 82: NOx at end of post-flame region vs. turbulent mixing time (flame zone residence time = 3.948 ms)
Figure 83: Temperatures at the end of post-flame region vs. equivalence ratio (at flame zone residence time = 3.948 ms)

Figure 84: NO$_x$ at the end of post-flame region vs. equivalence ratio (at flame zone residence time = 3.948 ms)
4.2.3.1.2. **PSR Limit**

As discussed before, a perfectly-stirred reactor (PSR) model assumes perfect mixing inside the reactor volume, and if used to model a flame zone, it would imply an infinite mixing frequency and zero mixing time. If compared to a PaSR, the PSR would serve as a limit when mixing time is decreased. To check the proper application of a PaSR, the values from the PSR should match an asymptotic value at an infinitesimal mixing time. To check that limit, temperatures and NO\textsubscript{x} values are also calculated using a PSR model for the flame zone (hence the reactor network for the flame would be PSR+PFR), and results are also added to the previous plots. PSR results serve as both:

1. a limit to check the PaSR results, and
2. a comparison to the usually selected method of PSR and PFR to model the flame and post-flame zones, respectively in literature, e.g. Elkady et al. [82].

As a check for the PaSR model, it is clear from Figure 81 through Figure 84 that values using a PaSR for the flame zone approach those using a PSR for the flame zone as mixing time tends to zero.

4.2.3.1.3. **PFR Limit**

As mixing time increases (mixing frequency decreases), the reactants would mix less with hot intermediates and products of combustion, which would make the flame harder to stabilize and flame could then not be sustained. Therefore, after the mixing time increases beyond a certain limit, the flame would blow off, and the results of a PaSR simulation would
result in the same reactant temperature and composition. If the PaSR mixing time increases to infinity, there is no axial mixing in the flame and this would correspond to a plug-flow reactor (PFR) model. To check the PFR limit, the PaSR simulation was repeated at increasing mixing times till the flame would blow off. The PaSR was also replaced by a PFR for the flame zone (hence the reactor network would be PFR+PFR), and calculations were performed again. As can be seen in the previous figures, as mixing time increases, the flame would blow off. After that mixing time, all simulations of a PaSR would correspond to a PFR, the product temperature would be equal to the reactants temperature, and NO\textsubscript{x} emissions would basically be zero. The same blowout phenomenon is also observed and discussed in [90].

If the flame zone residence time is decreased beyond a certain limit, the flame would also blow off, because the flame volume would not be enough for the reactants to burn before exiting the flame zone. This could occur if the chemical time scales are less than the flame residence time, which also happens for PSR models, as discussed before in chapter 3.

4.2.3.1.4. Attempts for Considering Recirculation

It is worth mentioning that an attempt was made to add a PSR reactor to the reactor network that accounts for recirculation around the flame to facilitate stabilization; Figure 85. This reactor was to be added such that the flame zone volume is divided to the forward-flow flame zone and the recirculation flame zone. However, because the sum of volumes of both zones is constant, this was not useful as the residence time of either zone was not large enough to avoid blow off, at the mixing times that correspond to the present case study. Another disadvantage of adding such recirculation zone was the significant increase of simulation hours by more than one order of magnitude, as tens of iterations were required to reach a
thermochemical balance for the flame zone loop, if the flame recirculation zone was modeled by a PSR. Simulation hours would almost multiply if a PaSR would be used instead; Figure 86.

![Diagram 1](image1.png)

**Figure 85:** Attempt for adding recirculation to the modeling chemical reactor network (using PSR for the recirculation zone)

![Diagram 2](image2.png)

**Figure 86:** Attempt for adding recirculation to the modeling chemical reactor network (using PaSR for the recirculation zone)

**4.2.3.2. Effect of Total Residence Time**

As the post-flame (PFR) region residence time increases, as shown in Figure 87 and Figure 88, flame temperature is almost constant after the transition from the flame zone (PaSR) to the post-flame zone (PFR), with a value nearly equal to the combustion equilibrium temperature. On the other side, because NO\textsubscript{x} formation is slow, and reaches equilibrium in a residence time much larger than typical gas turbine combustor residence times, NO\textsubscript{x} level has an approximately linear trend and is proportional to the residence time in the post-flame zone, as
shown in Figure 89 and Figure 90. The linear trend of NOx with total residence time was used to extrapolate Leonard and Correa’s work [81] for validation with the present study. The dotted segments refer to what occurs inside the flame zone (PaSR) since the values are assumed to jump suddenly from the inlet unburnt conditions to the PaSR values (because PaSR is a 0-D model).

Figure 87: Effect of total residence time on flame temperatures for turbulent mixing time of 0.01 ms and flame zone residence time of 2.632 ms
Figure 88: Effect of total residence time on flame temperatures for turbulent mixing time of 0.50 ms and flame zone residence time of 2.632 ms

Figure 89: Effect of total residence time on NO\textsubscript{x} for turbulent mixing time of 0.01 ms and flame zone residence time of 2.632 ms
4.2.3.3. Effect of Flame Zone Residence Time

Results at flame zone residence times of 2.632 ms, 3.948 ms and 5.264 ms (corresponding to flame heights of 0.6”, 0.9” and 1.2”) were studied. As shown in the example Figure 91 at $\Phi = 0.66$, flame zone residence time has a slight effect on NO$_x$ emissions. As flame zone residence time increases, NO$_x$ inside the flame slightly increases (since the range of flame zone residence time variation is small). However, since the total residence time is fixed, the residence time of the flame zone increases on the expense of that of the post-flame zone, which negatively affects the buildup of NO inside the post-flame region. Hence the net effect can be either a slight increase or decrease of NO$_x$ with increasing flame zone residence time. As can be
seen in Figure 91, the effect is negligible.

One might suggest that NO\textsubscript{x} levels be reduced by decreasing the turbulent mixing inside a combustor in lean premixed combustion. However, stabilizing the flame becomes more and more difficult, as demonstrated by the flame blow-off involved with large mixing times.

4.3. Carbon Monoxide (CO) Emissions

For convenience, CO levels are also calculated both experimentally and using chemical kinetics modeling. It is noted from Figure 92 that the chemical kinetics modeling network overestimated CO by about one order of magnitude. Such overestimation of CO is also
concluded in literature using PSR+PFR as in the work by Elkady et al. [82]. CO spikes at lean blowout were not observed as the equivalence ratio was not varied slowly near lean blowout conditions.

Figure 92: CO level comparison between experimental and modeling results

The effect of mixing time on CO is shown in Figure 93. As mixing time increases, CO slightly decreases due to less mixing between radicals and intermediates. The effect is slight because CO formation is much faster than NO formation. It can reach equilibrium levels quickly, after it exits the flame section, as seen in its dependence on total residence time in Figure 94.
Figure 93: Effect of turbulent mixing time on CO level at the end of the post-flame zone at flame zone residence time of 3.948 ms

Figure 94: Effect of total residence time on CO level for a mixing time of 0.50 ms and flame zone residence time of 3.948 ms
4.4. Summary and Conclusions

1. Two perforated-plate nozzle configurations were used to investigate effect of turbulence on NOx emissions from lean perfectly-premixed methane/air combustion at a range of equivalence ratio range of 0.54 to 0.85, at a combustor pressure of 5 atm, and inlet preheated temperature of 495 K.

2. Due to difference in nozzle geometry, the “turbulent” 48T0110 nozzle was found to deliver turbulence levels about 2-3 times those from the “normal” 48N0110 nozzle, with the same jet velocity. For the same nozzle, turbulence levels were found to increase with equivalence ratio due to flame-generated turbulence and combustion instabilities.

3. Change of NOx levels between the nozzles was not significant at different equivalence ratios. That can also be concluded using a chemical kinetics (PaSR + PFR) model.

4. Modeling shows that NOx emissions increase with decreasing mixing times, due to the higher flame zone temperatures and oxygen atom levels, because of the higher mixing rate of fresh reactants with hot intermediates and between intermediates and hence increasing temperatures and enhancing chemical reactions.

5. Although decreasing mixing length increases temperatures inside the flame zone, the effect on temperatures in the post-flame zone is negligible, as temperature quickly reaches equilibrium values after the flame zone.

6. NOx emissions increase with total residence time in a linear trend, due to the constant rate of formation of NO, as temperatures and oxygen levels inside the post-flame zones are constant at equilibrium values.

7. NOx formation is attributed to the buildup in the flame and post-flame zones with different contributions. The difference is mainly due to different rates of formation in the
flame zone.

8. Flame zone residence time has negligible effect on NO$_x$, in the range of the present study.

9. Modeling the flame as a PaSR + PFR (as well as PSR + PFR) overestimates CO levels by about one order of magnitude, and chemical kinetic models are still not suitable for CO prediction. However, chemical kinetic modeling gives acceptable values for NO$_x$ prediction.
Appendix A

Flame Temperature Correction for Heat Losses

In order to calculate heat losses from the combustor due to window air cooling, a heat transfer network is constructed as shown in the figure 24.

As shown above heat losses are calculated from balancing contributions of heat transfer by different modes inside the combustor (flame and gases side) and outside the combustor (cooling side), in form of radiation, convection and conduction. Such balance is formulated as

\[
Q_{\text{loss}} = Q_{\text{conv}, g} + Q_{\text{rad}, g} + Q_{\text{cond along wall}} = Q_{\text{conv}, c} + Q_{\text{rad}, c} = Q_{\text{cond}}
\]  

(54)

This balance can be reformulated as

\[
(R_1 + C_1 + K)\Delta A_g = (R_2 + C_2)\Delta A_c = K_{1-2}\Delta A_g
\]  

(55)

where \(R_1\) is the radiation heat flux from combustion gases and flame, \(R_2\) is the radiation heat flux to cooling air in the surrounding vessel, \(C_1\) is the convection heat flux from combustion gases, \(C_2\) is the convection heat flux to the cooling air in the surrounding vessel, \(K\) is the heat conduction along the liner wall, \(K_{1-2}\) is the conduction heat flux through wall, and \(\Delta A_g\) and \(\Delta A_c\) are the inside and outside surface areas, respectively.

Ignoring the conduction heat flux along the combustor liner \(K\) and ignoring combustor thickness (i.e. \(\Delta A_g \approx \Delta A_c\)). Hence,
Different contributions of heat losses are calculated as follows:

1. **Internal Radiation**

For the combustion gases, the total emitted radiation is composed of nonluminous radiation (that emanates from certain heteropolar gases, notably CO$_2$ and water vapor) and luminous radiation (that is due to emission from soot particles in the flame). To account for both, the internal radiation heat flux can be formulated as:

\[
R_1 = 0.5\sigma(1 + \varepsilon_w)\varepsilon_g T^1.5_{gases,\text{avg}} \left(T^2.5_{gases,\text{avg}} - T^{2.5}_{\text{liner,avg}}\right)
\]  

(57)

where \(\sigma\) is Stefan Boltzmann constant, \(\varepsilon_w\) is the wall emissivity (here for stainless steel \(\sim 0.83\)), \(\varepsilon_g\) is the gas emissivity at gas temperature \(T_{gases,\text{avg}}\), and is given by

\[
\varepsilon_g = 1 - \exp\left(-290PL(qL_b)^{0.5}T^{-1.5}_{gases,\text{avg}}\right)
\]  

(58)

where \(P\) is the combustion pressure (here = 5 atm), \(L\) is the luminosity factor taken as unity for lean premixed flames, \(q\) is the fuel/air ratio by mass and \(L_b\) is the beam length determined by the size and shape of the gas volume, and can be estimated from

\[
l_b = 3.4 \frac{\text{volume}}{\text{surface area}}.
\]  

(59)

2. **External Radiation**

The radiation heat transfer \(R_2\) from the liner wall to the cooling vessel casing can be approximated by assuming gray surfaces with emissivities \(\varepsilon_w\) and \(\varepsilon_c\) (here for stainless steel \(\sim 0.83\)) and assuming that \(T_{\text{liner,avg}}\) and \(T_{\text{cooling,avg}}\) are uniform in the axial direction. The external radiation heat transfer can be calculated as

\[
R_2 = \frac{\sigma}{\varepsilon_c + \varepsilon_w(1-\varepsilon_c)} \frac{\varepsilon_w \varepsilon_c}{\text{combustor side length}} \frac{\text{vessel side length}}{\left(T_{\text{liner,avg}}^4 - T_{\text{vessel,avg}}^4\right)}
\]  

(60)

The average vessel temperature is approximated as

\[
T_{\text{vessel,avg}} \approx 0.5\left(T_{\text{cooling,avg}} + T_\infty\right)
\]  

(61)

where \(T_\infty\) is the surrounding temperature, and \(T_{\text{cooling,avg}}\) is approximated as

---

3. **Internal Convection**

Convection heat flux from the combustion gases can be estimated as

\[
C_1 = 0.020 \frac{k_g}{d_{h1}^{0.2}} \left( \frac{\dot{m}_g}{A_g \mu_g} \right)^{0.8} (T_{gases,avg} - T_{liner,g})
\]

where \(d_{h1}\) is the hydraulic diameter of the combustor (= combustor side length), \(A_g\) is the combustor flow area (= (combustor side length)^2), \(\dot{m}_g\) is the flow rate of combustion gases, \(k_g\) is the combustion gases conductivity, and \(\mu_g\) is the combustion gases viscosity.

The properties \(k_g\) and \(\mu_g\) can be estimated by the formula

\[
\mu_g \approx \sum_{i=1}^{n} \frac{x_i \mu_i}{\sum_{j=1}^{n} x_j \phi_{ij}}
\]

where \(x_i\) is the molar fraction of component \(i\) (calculated from combustion gas composition by FTIR measurements), \(\mu_i\) is the viscosity of component \(i\) at \(T_{gases,avg}\), and

\[
\phi_{ij} = \left[ 1 + \left( \frac{\mu_i}{\mu_j} \right)^{1/2} \left( \frac{M_j}{M_i} \right)^{1/4} \right]^2 \left( 8 + 8M_i/M_j \right)^{1/2}
\]

\(M_i\) and \(M_j\) are the molecular weights of components \(i\) and \(j\), respectively.

Similar calculation of \(k_g\) can be performed if \(\mu_i\) is replaced by \(k_i\).

4. **External Convection**

From the dependence of heat transfer coefficient on Reynolds number and using proper correlations, the external convection heat flux \(C_2\) can be calculated as

\[
C_2 = 0.020 \frac{k_a}{D_{vessel}^{0.2}} \left( \frac{\dot{m}_a}{A_a \mu_a} \right)^{0.8} (T_{liner,c} - T_{cooling,avg})
\]

where

\[
D_{vessel} = \frac{\text{vessel side length}^2 - \text{combustor side length}^2}{\text{combustor side length}}
\]

is the hydraulic diameter of the cooling vessel, \(A_a = \text{vessel side length}^2 - \text{combustor side length}^2\), is the vessel flow area, \(\dot{m}_a\) is the flow rate of cooling air, \(k_a\) is the cooling air conductivity, and \(\mu_a\) is the cooling air viscosity.

---

Using the combustor operating conditions and component dimensions, the above equations were implemented to an iteration algorithm and was solved by Engineering Equation Solver (EES)\textsuperscript{11} for heat losses $Q_{\text{loss}}$.

After heat losses were calculated, adiabatic flame temperatures were corrected to account for heat losses, using a one-dimensional model as shown in figure 25.

\begin{equation}
T_{\text{flame, corrected}} = T_{\text{flame, adiabatic}} - \frac{Q_{\text{loss, jet}}}{m_{\text{tot, jet}} \cdot c_{p, \text{gases}}}
\end{equation}

\textbf{Figure 25 (revisited): Corrected temperature calculation procedure}

\textsuperscript{11} Engineering Equation Solver (EES), F-Chart Software. \url{http://www.fchart.com/ees/}
Appendix B

Dimensional Drawing for Emission Sampling Probe

Detail A
Probe Tip

Detail B
1/2" To 3/4" NPT Adapter for Fixation in Combustor

Detail C
3/8" To 1/2" Reducer

Cooling Water Out

Detail D
3/8" Tee

Detail E
1/4" Tee

Detail G
1/4" To 1/8" Reducer

To Emission Analysis Station
Detail B

1/2” to 3/4” NPT Adapter for Fixation in Combustor
**Detail C**

3/8” to 1/2” Reducer

**Detail D**

3/8” Tee for Cooling Water Exit
Detail E

3/8” to 1/4” Reducer

Detail F

1/4” Tee for Cooling Water Inlet
Detail G

1/4” to 1/8” Reducer
References


[92] Hamdi, M., Bentīcha, H., and Sassi, M., "Influence of Turbulent Mixing on Flameless Combustion and NO\textsubscript{x} Formation."


