Characterisation of oxy-fuel flames using laser based diagnostics techniques

by

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Declaration

I herewith certify that all material in this dissertation which is not my own work has been properly acknowledged.

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Abstract

Oxygen enhanced oxy-fuel laminar and turbulent flame structure in a co-flow non-premixed jet burner are investigated. The measurement of intermediate species such as hydroxyl (OH) and formaldehyde (CH$_2$O) and temperature are the focus of this work. The species concentrations were measured using planar laser induced fluorescence (PLIF) and the temperature using Rayleigh scattering. ‘Traditional Rayleigh’ requires a constant Rayleigh cross-section throughout the combustion process. This is impossible in high temperature oxy-fuel flames due to thermal decomposition. Derived temperature from Rayleigh signals is hence prone to inaccuracy.

A direct comparison of measured and numerically-calculated Rayleigh signals can eliminate this error. Numerical Rayleigh signals are relatively easily calculated with knowledge of temperature and species concentration. The feasibility of adopting this procedure to validate the numerical model was investigated in laminar and turbulent flames. Sensitivity studies including radiation models, chemical kinetics mechanisms and the Soret effect were performed in laminar flames.

Another Rayleigh technique, polarised/ depolarised Rayleigh was employed in a joint temperature, OH and CH$_2$O measurement. The effect of varying O$_2$ and jet Reynolds number on the flame structure was investigated. The applicability of determining heat release rate (HRR) using the product of [OH]x[CH$_2$O] was also determined. [OH]x[CH$_2$O] and HRR showed good spatial correlation in the main oxidation zone, but underestimated HRR in the secondary oxidation zone.

Finally, analysis of thermal diffusion structures using high resolution polarised/ depolarised Rayleigh was performed. The analysis revealed the thickness of the diffusion layer is proportional to the temperature, axial location and O$_2$ concentration. Increase of Reynolds number, however, reduces layer thickness.

In summary, this work has used a suite of optical diagnostics to make the first structural survey of high temperature oxy-fuel flames, starting with overall flame shape through macroscopic localised extinction to microscopic thermal diffusion.
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# Contents

1 Introduction........................................................................................................................................15  
   1.1 Motivation ......................................................................................................................................15  
   1.2 Objectives and approach ...............................................................................................................16  
   1.3 Thesis structure ............................................................................................................................17  
2 Literature review ..................................................................................................................................19  
   2.1 Chapter overview ..........................................................................................................................19  
   2.2 Fundamentals of non-premixed flames .........................................................................................19  
      2.2.1 Laminar non-premixed flames ............................................................................................21  
      2.2.2 Turbulent non-premixed flames .........................................................................................23  
      2.2.3 Inverted flames ....................................................................................................................25  
   2.3 Role of laser diagnostics in combustion ......................................................................................27  
      2.3.1 Temperature measurement ..................................................................................................28  
      2.3.2 Concentration measurement ...............................................................................................31  
      2.3.3 Multi-scalar measurements .................................................................................................32  
   2.4 Oxy-fuel flames overview ..........................................................................................................34  
      2.4.1 Differential diffusion effect ................................................................................................34  
      2.4.2 Soot formation and flame radiation ....................................................................................35  
      2.4.3 Flame chemistry .................................................................................................................36  
      2.4.4 Flame speed and stability ....................................................................................................38  
3 Experimental techniques and equipment .........................................................................................40  
   3.1 Background on measurement techniques ...................................................................................40  
      3.1.1 Hot wire anemometry ..........................................................................................................40  
      3.1.2 Rayleigh scattering ..............................................................................................................41  
      3.1.3 Laser Induced Fluorescence ...............................................................................................44  
         3.1.3.1 OH-PLIF .......................................................................................................................46  
         3.1.3.2 CH$_2$O-PLIF ...............................................................................................................48  
   3.2 Experimental equipment .............................................................................................................49  
      3.2.1 Burner and flow configuration ..............................................................................................49
3.2.2 Lasers.................................................................................................................. 51
  3.2.2.1 Nd:YAG lasers ............................................................................................... 51
  3.2.2.2 Dye laser ....................................................................................................... 53
3.2.3 Light detectors..................................................................................................... 54
  3.2.3.1 CCD cameras .............................................................................................. 55
  3.2.3.2 Intensified CCD cameras ............................................................................. 56
4 Simultaneous Rayleigh and OH-PLIF signals ....................................................... 58
  4.1 Introduction ........................................................................................................ 58
  4.2 Methodology ....................................................................................................... 61
    4.2.1 Boundary conditions ....................................................................................... 61
    4.2.2 Fuel and oxidiser selection ............................................................................ 62
    4.2.3 Experimental setup ....................................................................................... 66
    4.2.4 Post processing ............................................................................................. 67
  4.3 Numerical Approach ........................................................................................... 69
  4.4 Results and discussion ....................................................................................... 71
    4.4.1 Comparison of experimental and numerical Rayleigh signals in laminar flame. 72
    4.4.2 Comparison of experimental and numerical Rayleigh signals in turbulent flame 76
    4.4.3 Comparison of normal and inverted flame structure using Rayleigh and OH-PLIF signals 82
  4.5 Uncertainty analysis ........................................................................................... 84
  4.6 Conclusion ......................................................................................................... 85
5 Simultaneous mixture fraction, temperature, OH- and CH₂O-PLIF measurement...... 88
  5.1 Introduction ........................................................................................................ 88
  5.2 Methodology ....................................................................................................... 89
    5.2.1 Fuel and oxidiser selection ............................................................................ 89
    5.2.2 Experimental setup ....................................................................................... 94
    5.2.3 Post processing ............................................................................................. 97
      5.2.3.1 Polarisation and depolarisation post processing ........................................ 98
      5.2.3.2 OH-PLIF post processing ....................................................................... 101
      5.2.3.3 CH₂O post processing .......................................................................... 102
5.3 **Result and discussion** ........................................................................................................ 105
   5.3.1 Instantaneous, mean and RMS of temperature, OH and CH$_2$O signals analysis. 105
   5.3.2 Further analysis on pocket OH and CH$_2$O ................................................................. 114
   5.3.3 Reaction zone thicknesses of OH ................................................................................... 118
   5.3.4 Scatter plot of temperature and localised extinction analysis ........................................ 120
5.4 **Uncertainty analysis** ........................................................................................................ 124
5.5 **Conclusion** ....................................................................................................................... 125

6 **Analysis on the applicability of heat release represented by [OH][CH$_2$O] in non-premixed oxy-fuel flames** .................................................................................................................. 128
   6.1 **Introduction** .................................................................................................................. 128
   6.2 **Methodology** ................................................................................................................. 129
      6.2.1 Temperature dependence of the LIF signals .............................................................. 129
      6.2.2 Data reduction procedures ......................................................................................... 130
   6.3 **Result and discussion** .................................................................................................. 131
      6.3.1 The structure of the heat release profile .................................................................... 131
      6.3.2 Comparison of simulated and measured OH, CH$_2$O and [OH][CH$_2$O] quantities 135
      6.3.3 Instantaneous temperature, OH, CH$_2$O, gradient-squared temperature and reaction rate images .............................................................................................................................. 137
      6.3.4 Scatter plots of FRR vs temperature and FRR vs gradient-squared temperature 139
   6.4 **Conclusion** .................................................................................................................... 142

7 **Spatial analysis of dissipative structure in oxy-fuel flames using high-resolution Rayleigh** .......................................................................................................................... 144
   7.1 **Introduction** ................................................................................................................. 144
   7.2 **Methodology** ................................................................................................................ 146
      7.2.1 Experimental setup .................................................................................................... 146
      7.2.2 Data processing ......................................................................................................... 147
   7.3 **Result and discussion** .................................................................................................. 149
      7.3.1 Mean contribution of $\nabla T^2$ and $\nabla T^2$ ................................................................. 149
List of Tables

TABLE 3-1: SPECIFICATIONS OF IROS USED IN THE EXPERIMENT .......................................................... 57
TABLE 4-1: EXPERIMENTAL CONDITIONS FOR RAYLEIGH AND SIMULTANEOUS RAYLEIGH AND OH-PLIF
MEASUREMENT ....................................................................................................................................... 67
TABLE 5-1: EXPERIMENTAL CONDITIONS FOR SIMULTANEOUS MIXTURE FRACTION, TEMPERATURE, OH- AND
CH₂O-PLIF MEASUREMENT .................................................................................................................... 97
TABLE 5-2: MEAN LAYER THICKNESS OF OH REACTION ZONE .................................................................. 120
TABLE 6-1: LOCAL EXTINCTION PERCENTAGE (NUMBER OF PIXELS WITH FRR=0 TO THE NUMBER OF SAMPLED
PIXELS) IN 50 % AND 55 % O₂ FLAMES AT DIFFERENT REYNOLDS NUMBER, AT X/D=5 AND X/D=10 ...... 141
TABLE 7-1: TEST CASES FOR THE HIGH-RESOLUTION RAYLEIGH TEMPERATURE MEASUREMENTS ........... 147
List of Figures

FIGURE 2-1: ILLUSTRATION OF THE APPEARANCE OF FLAMES. AS THE JET REYNOLDS NUMBER INCREASES, LAMINAR FLAMES BECOME LONGER, UNTIL THEY TRANSITION INTO A TURBULENCE-DOMINATED MODE WHERE FLAME LENGTH IS NO LONGER PROPORTIONAL TO RE. ADAPTED FROM WARNATZ ET AL [2] .... 20

FIGURE 2-2: ILLUSTRATION OF LAMINAR NON-PREMIXED FLAME. ADAPTED FROM TURNS [1]. THE LEFT GRAPHS ARE THE TEMPERATURE PROFILES, WHILE ON THE RIGHT ARE THE MASS FRACTION PROFILES OF FUEL, OXIDISER AND PRODUCTS AT DIFFERENT AXIAL POSITIONS. ....................................................... 22

FIGURE 2-3: ILLUSTRATION OF VORTEX GENERATION IN TURBULENT NON-PREMIXED FLAME. ADAPTED FROM RENARD [4]. ........................................................................................................ 23

FIGURE 2-4: ILLUSTRATION OF ENERGY STATES IN INELASTIC RAMAN SCATTERING PROCESSES SHOWING STOKES AND ANTI-STOKES PHOTONS AND COMPARISON TO RAYLEIGH SCATTERING. ADAPTED FROM WARNATZ [2]. ........................................................................................................ 31

FIGURE 3-1: ENERGY LEVEL DIAGRAM FOR A TWO-LEVEL FLUORESCENCE MODEL ADAPTED FROM [5] .......... 45

FIGURE 3-2: BOLTZMANN FRACTION POPULATION FOR Q(6), Q(7), Q(8), AND Q(9) ABSORPTION LINES AT TEMPERATURE OF 1000 – 2500 K ................................................................. 47

FIGURE 3-3: ABSORPTION SPECTRUM OF OH (A²Σ⁺-X²Π) AT 2000 K, SIMULATED USING LIFBASE ............... 48

FIGURE 3-4: A SCHEMATIC DIAGRAM OF THE NON-PREMIXED JET BURNER (A) AND A PHOTO OF THE BURNER SECURED TO AN OPTICAL TABLE AND PLACED ON TOP OF A LABORATORY JACK (B). ......................... 49

FIGURE 3-5: ILLUSTRATION OF EQUIPMENT AND GAS LINES ARRANGEMENT FOR EXPERIMENT IN DISCUSSED CHAPTER 4 ........................................................................................................ 50

FIGURE 3-6: ILLUSTRATION OF THALES MULTI-YAG LASER SYSTEM IN 532 NM AND 355 NM CONFIGURATION USED IN CHAPTER 5 ........................................................................................................ 52

FIGURE 3-7: QUANTUM EFFICIENCY VS WAVELENGTH OF VARIOUS PHOTOCATHODE ADAPTED FROM [3]. SBA IS SUPER BIALKALI AND UBA IS ULTRA BIALKALI ...................................................... 57

FIGURE 4-1: COMPARISON OF MEAN AND RMS AXIAL VELOCITY FROM THE HOT WIRE MEASUREMENT AND NUMERICAL CALCULATION AT RE=18000 AT 1,3,10 AND 20 MM ABOVE THE NOZZLE TIP ............. 61

FIGURE 4-2: NORMALISED RAYLEIGH CROSS SECTION (TOP LEFT), NORMALISED RAYLEIGH SIGNAL (TOP RIGHT), MOLE FRACTION AT 1000 S⁻¹ (BOTTOM LEFT) AND TEMPERATURE (BOTTOM RIGHT) FROM THE LAMINAR OPPOSED FLAME CALCULATION AT DIFFERENT STRAIN RATES FOR (FUEL: 17.5 % CH₄, 40 % CO₂, 42.5 % H₂, OXIDISER: 68 % O₂, 32% CO₂) ........................................................................................................ 63

FIGURE 4-3: ILLUSTRATION OF SIMULTANEOUS RAYLEIGH AND OH-LIF MEASUREMENT SETUP .................. 65

FIGURE 4-4: COMPARISON OF RADIATION MODELS TO THE EXPERIMENTAL RESULT AT TWO DIFFERENT RADIAL LOCATIONS (TOP – 60-85 MM, BOTTOM – 23-48 MM): (A) EXPERIMENT (B) ADIABATIC (C) OTM (D) P1. COMPARISON OF RADIATION MODELS ALONG A RADIAL LINE AT 35 MM (E) AND 75 MM (F) ............... 73

FIGURE 4-5: COMPARISON OF EXPERIMENTAL AND NUMERICAL SIMULATION WITH AND WITHOUT SORET EFFECT (A) EXPERIMENTAL (B) P1 SIMULATION WITH SORET EFFECT (C) P1 SIMULATION WITHOUT SORET

10
EFFECT. COMPARISON OF EXPERIMENTAL AND SIMULATION WITH AND WITHOUT SORRET EFFECT ALONG A LINE AT (D) 35 MM AND (E) 75 MM. ............................................................................................................................ 75

FIGURE 4-6 : SINGLE SHOT COMPARISON (FOR QUALITATIVE ILLUSTRATION) OF TURBULENT FLAME AT RE=18,000, LEFT (EXPERIMENT) AND RIGHT (SIMULATION). BOTTOM (23 MM – 48 MM), MIDDLE (48 MM-73 MM) AND TOP (98 MM – 123 MM). ............................................................................................................................ 76

FIGURE 4-7: COMPARISON OF THE (A) MEAN AND (B) VARIANCE OF THE RAYLEIGH RATIO FROM EXPERIMENT AND NUMERICAL SIMULATION. THE EXPERIMENT IS ALWAYS ON THE LEFT, WHILE NUMERICAL ON THE RIGHT. ............................................................................................................................ 79

FIGURE 4-8: COMPARISON OF THE EXPERIMENTAL AND NUMERICAL DATA: MEAN (LEFT) AND RMS RAYLEIGH RATIO (RIGHT) ............................................................................................................................ 80

FIGURE 4-9: PDF OF RAYLEIGH RATIO: USING DATA AT THE LOCATION OF THE PEAK IN THE MEAN PROFILE (LEFT) AND DATA FROM LOCATIONS WHICH SHOW STRONGEST FLUCTUATIONS (RIGHT) .............. 81

FIGURE 4-10: COMPARISON OF NORMAL AND INVERTED FLAMES FOR EXPERIMENTAL AND NUMERICAL MEAN AND RMS RAYLEIGH RATIO SIGNAL (LEFT) AND OH-PLIF (RIGHT). ............................................................................................................................ 83

FIGURE 4-11: COMPARISON OF THE PDF AT DIFFERENT RADIAL POSITIONS OF THE RAYLEIGH RATIO RMS PEAK, OH-PLIF MEAN PEAK AND OH-PLIF RMS PEAK AT HEIGHT OF 67 MM FROM NOZZLE EXPERIMENTAL DATA (TOP) AND NUMERICAL DATA (BOTTOM) FOR IDF FLAME ...................................................................................... 83

FIGURE 5-1: SPECIES AND TEMPERATURE PROFILES OF FUEL: 55 % H₂, 45% CH₄, OXIDISER: 50 % O₂, 50 % CO₂ CALCULATED USING CHEMKIN EMPLOYING OPPOSED JET LAMINAR FLAME, (A) A=100 S⁻¹ AND 1000 S⁻¹, EQUAL DIFFUSIVITY (B) A=100 S⁻¹ AND 1000 S⁻¹, MULTI-COMPONENT. TEMPERATURE, NORMALISED POLARISED AND DEPOLARISED RAYLEIGH CROSS-SECTION AT DIFFERENT STRAIN RATE (A=100 S⁻¹ – 1000 S⁻¹) USING (C) EQUAL DIFFUSIVITY (D) MULTI-COMPONENT TRANSPORT. ...................................................................................... 89

FIGURE 5-2: (A) PREDICTED POLARISED AND DEPOLARISED RAYLEIGH RATIO, SIGNAL DIFFERENCE AND SIGNAL DIFFERENCE USING NON-LINEAR COMBINATION OF POLARISED AND DEPOLARISED RAYLEIGH SIGNAL CALCULATED AT STRAIN RATE OF 800 S⁻¹, (B) NON-LINEAR COMBINATION RAYLEIGH SIGNALS AT DIFFERENT STRAIN RATES (100 S⁻¹ – 1000 S⁻¹ (TOP) AND THE PERCENTAGE DIFFERENCE OF THE RAYLEIGH SIGNALS (BOTTOM). ............................................................................................................................ 92

FIGURE 5-3: EXPERIMENT SETUP FOR SIMULTANEOUS MIXTURE FRACTION, TEMPERATURE, OH- AND CH₂O-PLIF MEASUREMENT ............................................................................................................................ 94

FIGURE 5-4: POLARISATION AND DEPOLARISATION RAYLEIGH IMAGES AT EVERY PROCESSING STAGE: (A) LEFT: INITIAL POST-PROCESSED POLARISED, RIGHT: INITIAL POST-PROCESSED DEPOLARISED (B) LEFT: NORMALISED POLARISED, RIGHT: NORMALISED DEPOLARISED (C) SIGNAL DIFFERENCE (D) MIXTURE FRACTION (E) NORMALISED RAYLEIGH CROSS SECTION (F) CORRECTED TEMPERATURE (G) UNCORRECTED TEMPERATURE ............................................................................................................................ 100

FIGURE 5-5: TEMPERATURE SENSITIVITY ANALYSIS AT DIFFERENT STRAIN RATE (A) TEMPERATURE VS RADIAL LOCATION FROM THE NOZZLE CENTRE, (B) TEMPERATURE VS MIXTURE FRACTION ...................................................................................... 101

FIGURE 5-6: COMPARISONS OF UNCORRECTED AND UNCORRECTED CH₂O SIGNALS ............................................................................................................................ 103
FIGURE 5-7: COMPARISON OF NORMALISED CH$_2$O SIGNALS BETWEEN RAW, ADAPTING QUenchING MODELS AND NUMERICAL SIMULATION ........................................................................................................... 104

FIGURE 5-8: SELECTION OF INSTANTANEOUS TRIPLET IMAGES (LEFT) OF TEMPERATURE, OH AND CH$_2$O AT DIFFERENT REYNOLDS NUMBER: RE=7000 (BOTTOM), RE=12000 (MIDDLE) AND RE=18000 (TOP) FOR 55 % O$_2$ FLAME. ON THE RIGHT IS THE MEAN (RED LINES) AND RMS (BLACK LINES) PROFILE OF TEMPERATURE, OH AND CH$_2$O AT 25 MM AXIAL LOCATION FOR DIFFERENT OXYGEN CONCENTRATION IN CO-FLOW. ............................................................................................................... 105

FIGURE 5-9: SELECTION OF INSTANTANEOUS TRIPLET IMAGES (LEFT) OF TEMPERATURE, OH AND CH$_2$O AT DIFFERENT REYNOLDS NUMBER: RE=7000 (BOTTOM), RE=12000 (MIDDLE) AND RE=18000 (TOP) FOR 55 % OXYGEN FLAME. ON THE RIGHT IS THE MEAN (RED LINES) AND RMS (BLACK LINES) PROFILE OF TEMPERATURE, OH AND CH$_2$O AT 50 MM AXIAL LOCATION FOR DIFFERENT OXYGEN CONCENTRATION IN CO-FLOW. ............................................................................................................... 108

FIGURE 5-10: SELECTION OF INSTANTANEOUS TRIPLET IMAGES (LEFT) OF TEMPERATURE, OH AND CH$_2$O AT DIFFERENT REYNOLDS NUMBER: RE=7000 (BOTTOM), RE=12000 (MIDDLE) AND RE=18000 (TOP) FOR 55 % OXYGEN FLAME. ON THE RIGHT IS THE MEAN (RED LINES) AND RMS (BLACK LINES) PROFILE OF TEMPERATURE, OH AND CH$_2$O AT 75 MM AXIAL LOCATION FOR DIFFERENT OXYGEN CONCENTRATION IN CO-FLOW. ............................................................................................................... 110

FIGURE 5-11: SELECTED SINGLE SHOT TRIPLET IMAGES OF TEMPERATURE, OH AND CH$_2$O OF 50 % O$_2$ FLAME AT RE=18000 AT 75 MM RADIAL POSITION ............................................................................................... 112

FIGURE 5-12: EXAMPLE OF LOCALISED EXTINCTION FROM 50 % O$_2$ FLAME AT 50 MM RADIAL POSITION. THE SINGLE SHOT IMAGES OF TEMPERATURE, OH AND CH$_2$O (TOP), JOINT PDF OF TEMPERATURE AND OH (MIDDLE) AND CH$_2$O AND TEMPERATURE (BOTTOM). ....................................................................... 114

FIGURE 5-13: EXAMPLE GENERATED POCKET CH$_2$O AND SURROUNDED BY OH FROM 50 % O$_2$ FLAME AT 50 MM RADIAL POSITION. THE SINGLE SHOT IMAGES OF TEMPERATURE, OH AND CH$_2$O (TOP), JOINT PDF OF TEMPERATURE AND OH (MIDDLE) AND CH$_2$O AND TEMPERATURE (BOTTOM). .................................................................... 115

FIGURE 5-14: EXAMPLE GENERATED RE-IGNITION PROCESS FROM 50 % O$_2$ FLAME AT 50MM RADIAL POSITION. THE SINGLE SHOT IMAGES OF TEMPERATURE, OH AND CH$_2$O (TOP), JOINT PDF OF TEMPERATURE AND OH (MIDDLE) AND CH$_2$O AND TEMPERATURE (BOTTOM). .................................................................... 116

FIGURE 5-15: EXAMPLE FINAL STAGE OF THE RE-IGNITION PROCESS FROM 50 % O$_2$ FLAME AT 50 MM RADIAL POSITION. THE SINGLE SHOT IMAGES OF TEMPERATURE, OH AND CH$_2$O (TOP), JOINT PDF OF TEMPERATURE AND OH (MIDDLE) AND CH$_2$O AND TEMPERATURE (BOTTOM) .................................................................... 117

FIGURE 5-16: PDF OF OH THICKNESSES FOR BOTH FLAMES AT RE=7000, RE=12000 AND RE=18000 AT DIFFERENT RADIAL LOCATION ........................................................................................................... 119

FIGURE 5-17: SCATTER PLOT OF TEMPERATURE AT X/D=5 (25 MM) AND X/D=10 (50 MM) FOR 50 % O$_2$ FLAME AT RE=12000 AND RE=18000. THE STOICHIOMETRIC MIXTURE FRACTION, F$_{ST}$ IS MARKED WITH GREY DOTTED VERTICAL LINE. ........................................................................................................... 120
FIGURE 5-18: PDF OF TEMPERATURE CONDITIONED ON NARROW BAND MIXTURE FRACTION (Δ=0.02) CENTRED AT HIGHEST GLOBAL AVERAGE TEMPERATURE FOR BOTH FLAMES AT RE=12000 AND RE=18000 AT DIFFERENT AXIAL POSITION. ........................................................................................................... 121

FIGURE 6-1: COMPARISON OF TOTAL HEAT RELEASE RATE AND MAIN REACTIONS CONTRIBUTED TO THE TOTAL HEAT RELEASE IN (A) PREMIXED CH₄-AIR (B) NON-PREMIXED CH₄-AIR (C) NON-PREMIXED OXY-FUEL FLAME OF FUEL: 55 % H₂, 45 % CH₄, OXIDISER: 55 % O₂, 45 % CO₂ AT STRAIN RATE OF 2500 s⁻¹ USING MULTI-COMPONENT TRANSPORT. A COMPARISON OF HEAT RELEASE AND FORWARD REACTION RATE OF [OH][CH₂O] FOR FUEL: 55 % H₂, 45 % CH₄, OXIDISER: 55 % O₂, 45 % CO₂ AT DIFFERENT STRAIN RATES AND LEWIS NUMBER IS SHOWN IN (D)........................................................................................................... 132

FIGURE 6-2: VARIATION OF THE FORWARD REACTION RATE (FRR) CONSTANT K(T) AT DIFFERENT STRAIN RATES AND RIGHT: SIMULATED NORMALISED HEAT RELEASE RATE (HRR) VS NORMALISED FORWARD REACTION RATE (FRR), WITH EQUIVALENCE PLOTTED AS A DASHED LINE........................................................................................................... 135

FIGURE 6-3: (LEFT) NORMALISED MEASURED SIGNALS OF LIF-OH, LIF-CH₂O AND FORWARD REACTION RATE. (RIGHT) CALCULATED OH AND CH₂O CONCENTRATIONS AND FORWARD REACTION RATE AT A=2500 s⁻¹. ............................................................................................................................................... 136

FIGURE 6-4: INSTANTANEOUS IMAGES OF TEMPERATURE, OH, CH₂O, GRADIENT-SQUARED TEMPERATURE AND REACTION RATE OF 50 % O₂ FLAME AT JET RE=18000 AT X/D=5. THE SIZE OF EACH IMAGE IS 8 MM X 28 MM, AND THE NOZZLE CENTRELINE IS MARKED WITH A VERTICAL WHITE DASHED LINE. THE REGION OF FORWARD REACTION RATE OF [OH][CH₂O] IS OVERLAPPED ON EACH IMAGE, REPRESENTED BY SOLID WHITE LINES. .................................................................................................................................. 138

FIGURE 6-5: SCATTER PLOT OF FORWARD REACTION RATE OF [OH][CH₂O] VS TEMPERATURE AT (A) RE=12000 AND (B) RE=18000 AND FORWARD REACTION RATE VS GRADIENT-SQUARED TEMPERATURE, Vᵀ² AT RE=12000 (C) AND RE=18000 (D).THE SCATTER PLOTS ARE FROM 50 % O₂ FLAME AT 25 MM. .................. 139

FIGURE 6-6: PROBABILITY DENSITY FUNCTION OF THE REACTION RATE INTENSITY FOR FLAME 50 % O₂ (A) AND 55 % O₂ (B). RE=12000 AND RE=18000 AT X/D=5 (25 MM) AND X/D=10 (50 MM) HEIGHT ABOVE BURNER ARE PLOTTED FOR EACH FLAME. .................................................................................................................. 140

FIGURE 7-1: STEP-BY-STEP POST-PROCESSING TO DETERMINE THE THERMAL DISSIPATION THICKNESS. (A) SINGLE-SHOT OF SMOOTHED TEMPERATURE IMAGE, (B) AVERAGE OF 500 TEMPERATURE IMAGES, (C) TEMPERATURE FLUCTUATION OF THE TEMPERATURE IMAGE (A), (D) SQUARED GRADIENT OF TEMPERATURE FLUCTUATION (C), (E) CENTRELINES OF THE LOCAL MAXIMA SQUARED GRADIENT OF TEMPERATURE FLUCTUATION (RED LINES), AND VISUALISATION OF PROFILES ALONG THE LAYER-NORMAL DIRECTION OF TEMPERATURE FLUCTUATION GRADIENT (YELLOW LINES) OF IMAGE (D), (F) ZOOMED IN OF THE PROFILES ALONG THE LAYER LOCAL MAXIMA OF TEMPERATURE GRADIENT. ........ 148

FIGURE 7-2: MEAN PROFILES OF SQUARED TOTAL GRADIENT, AXIAL AND RADIAL OF TEMPERATURE AND TEMPERATURE FLUCTUATIONS AT X/D = 5 AND X/D = 10 AT RE=12000 AND RE=18000 FOR 50 % O₂ FLAMES. Vᵀ², ∂Ṫ/∂x, ∂Ṫ/∂r ARE SHOWN IN RED LINES WHILE Vᵀ², ∂Ṫ/∂x, ∂Ṫ/∂r ARE SHOWN IN BLACK........................................................................................................................................ 150
FIGURE 7-3: PDF OF LAYER THICKNESSES CONDITIONED ON TEMPERATURE RANGE OF 300 K - 1700 K. EACH PDF LINE REPRESENTS A TEMPERATURE INTERVAL OF 200 K. ................................................................. 152

FIGURE 7-4: PDF OF SCALED LAYER THICKNESS OF 50 % O2 (TOP AND BOTTOM RIGHT) AND 55% O2 (BOTTOM LEFT) FLAMES AT X/D =5 AND X/D=10 .................................................................................................................. 153
1 Introduction

1.1 Motivation

Fossil fuels will remain an important energy source for decades to come. Fossil fuel combustion results, however, in increased atmospheric concentration of carbon dioxide (CO$_2$) and other greenhouse gasses$^1$. The current global level of CO$_2$ as of September 2016 is 400 parts per million (ppm) [6], 43 % higher compared to the pre-industrial era (about 280 ppm). Accumulated evidence that climate change is in large part due to this CO$_2$ emission has led to the establishment of several groups such as the Intergovernmental Panel on Climate Change (IPCC) which is responsible for providing a scientific view of the impact of current climate change to environmental and socio-economics, and the Kyoto Protocol which is a treaty that sets international limits and emission reduction targets.

A significant reduction of CO$_2$ could be achieved through adoption of one of several CO$_2$ Capture and Storage (CCS) technologies in combustion such as: pre-combustion capture, post-combustion capture and oxy-fuel combustion [7], to name a few. Oxy-fuel combustion stands out as the most promising and efficient, particularly in retrofitting existing coal-fired power plants [8] and natural gas fired gas turbines [9]. Oxy-fuel combustion is a concept where the air that is usually used in combustion, containing nitrogen (N$_2$), is replaced by pure oxygen (O$_2$). If a lower temperature flame is required, flue gas is added (usually rich in CO$_2$). The final product of this combustion will be CO$_2$ and water vapour, which can be separated through a condensation process enabling the CO$_2$ to be captured and stored or recycled [10]. High concentration of O$_2$ is usually used in start-up stage of furnace operation as flame stability can easily be achieved. Then, CO$_2$ is gradually increased to establish predetermined optimal combustion condition (e.g. 70 % CO$_2$ + 30 % O$_2$).

Though oxy-fuel combustion was proposed back in 1982 [11], the concept has only recently received significant attention from researchers in the combustion community. Thus, oxy-fuel knowledge is at an early stage and understanding being built from a fundamental level especially with regard to chemical aspects related to the combustion. At this stage, the

$^1$ Concentration of methane (CH$_4$), nitrous oxide (N$_2$O) and tropospheric ozone (O$_3$) in parts per billion (ppb) increased by about 154%, 22% and 42% respectively from mid-1800s to 2013 [6]
interplay between the experiment and numerical simulation is vital. The measured data, such as species concentration and temperature in a lab scale burner or combustor is often used to verify/validate the simulation result. For this reason, an accurate experimental measurement is indeed crucial. Laser diagnostics offer the capability make remote, non-intrusive, in-situ, spatially and temporally resolved measurements for the study of chemical and thermodynamic parameters in combustion. Several laser diagnostics techniques are available, and each has its own advantages and limitations.

Realising the need to enhance our knowledge in the oxy-fuel research area, this thesis is devoted to understanding and characterising oxy-fuel flames, focusing on oxygen enhanced systems and using the most suitable laser diagnostic techniques.

1.2 Objectives and approach

This thesis presents a study of oxygen-enhanced oxy-fuel flames diluted with carbon dioxide (CO₂) in a non-premixed jet burner. The main objective of the thesis is to understand the structure of oxy-enriched flames in laminar, turbulent normal diffusion flame (NDF) and turbulent inverted diffusion flame (IDF) arrangements. The data collected in this work includes the common quantities that are needed when validating numerical simulation codes, such as temperature and the intermediate species, hydroxyl (OH) and formaldehyde (CH₂O), using two-dimensional imaging techniques.

Owing to the complex nature of high temperature oxy-fuel flames in terms of a high concentration of intermediate species (O, H and OH) and the possibility of thermal decomposition of stable species (e.g. CO₂), a commonly used laser diagnostic technique for temperature measurement, namely Rayleigh scattering, is prone to inaccuracy. Hence, careful analysis and suitable strategies have to be used to tackle the challenges, in terms of reducing the experimental error as well as finding ways to compare the experimental and numerical data. These serve as secondary objectives of this thesis.

One strategy to reduce the experimental error is to directly compare the experimental measured signals to the numerically calculated signals. This way, the assumptions required to translate the measured signals into the physical quantities is avoided. The applicability of this approach will be analysed in laminar and turbulent non-premixed oxy-fuel flames.
side-by-side comparison in laminar flames enables a sensitivity study including radiation models, the Soret effect (thermophoresis) and reaction mechanisms suitable for oxy-fuel flames to be analysed. Further, a statistical comparison in turbulent flames is performed. A less complex fuel/oxidiser system in terms of the influence of molecular diffusivity will be used. A strict comparison between experiment and model in terms of the correlation of two quantities, i.e., a simultaneous scalars measurement, will also be performed.

More often, scalar quantities (e.g., temperature) rather than experimental signals are desired. Hence, another reliable technique needs to be explored to make this possible in oxygen-enhanced flames. A method, namely polarised/depolarised Rayleigh scattering will be presented, and a joint measurement of temperature, OH and CH$_2$O will be performed. Simultaneous measurement of these quantities enables further analysis of oxy-fuel flame structure including heat release, local extinction and thermal dissipation.

### 1.3 Thesis structure

Based on the outlined objectives and approach in the previous section, this thesis is structured as follows:

In chapter 2, a brief discussion of non-premixed flame fundamentals will be given, followed by a review of laser diagnostics applied to combustion, focusing on temperature and species measurements. To impart a basic understanding of the oxy-fuel flame to the reader, the last section is devoted to an overview of current progress in oxy-fuel flame studies.

Chapter 3 discusses in detail the laser diagnostic techniques that are used in this thesis, covering the theory of Rayleigh scattering, and planar laser induced-fluorescence (PLIF) of hydroxyl (OH-PLIF) and formaldehyde (CH$_2$O-PLIF). Equipment background is also given.

Chapter 4, 5, 6 and 7 are the results chapters. The results presented here are from different experiments, hence they are treated independently. Each has its own introduction, methodology, results and discussion sections. The chapters reveal progressively more detailed and fine-grained information about the structure of oxy-fuel flames.

The first experiment, which is used to validate a numerical simulation using simultaneous measurement of Rayleigh scattering and OH-PLIF in laminar and turbulent normal diffusion
flames (NDF) and inverted diffusion flames (IDF) is explained in Chapter 4. Experiment setup, fuel selection strategy, signal post-processing and a brief description on the numerical simulation approach are discussed. The applicability of the strategy in successfully comparing the experimental and numerical signals is then critically discussed in the results and discussion sections.

Chapter 5 describes the simultaneous measurement of mixture fraction, temperature, OH- and CH₂O-PLIF employing the polarisation/ depolarisation Rayleigh technique for mixture fraction and temperature measurement, and PLIF technique for the OH and CH₂O measurements. The fuel and oxidiser mixtures used in this chapter are different from Chapter 4, so the fuel and oxidiser selection process is also discussed. The complex experimental setup required to successfully run the joint measurement is explained in detail. Post-processing of the polarisation and depolarisation Rayleigh signals as well as OH and CH₂O signals comes next. The chapter ends with a results section, where the statistical results of temperature, OH and CH₂O are presented, focusing on the effects of varying experimental parameters (i.e. Reynolds number and O₂ concentration). The so-called “localised extinction” phenomenon, where the flame temporarily “goes out” is investigated.

Chapter 6 uses the same dataset discussed in Chapter 5. However, the analysis is significantly extended to investigate the applicability of measuring heat release by multiplying measured OH and CH₂O concentrations, [OH][CH₂O], in oxy-fuel flames. This provides a more clear physical link to the localised extinction phenomenon.

Chapter 7 shifts focus to a high resolution polarised/ depolarised Rayleigh temperature measurement, using the same fuel/oxidiser mixtures presented in Chapter 5 and 6 but a different optical setup. The data collected are used to resolve the fine structure of the flame and examine its thermal dissipation structure. The effect of varying Reynolds number and oxygen concentration on the thermal dissipation layer thickness are determined.

A brief conclusion, summarising the findings in this thesis is then presented in Chapter 8, followed by current research novelty and a discussion of the future direction this work could take.
2 Literature review

2.1 Chapter overview

This chapter starts with a discussion of the fundamentals of non-premixed flames which covers the flames investigated in this work. Then, relevant laser techniques are reviewed focusing on temperature and concentration as well as multi-scalar measurement. Finally, the oxy-fuel combustion research literature is summarised.

2.2 Fundamentals of non-premixed flames

This work was only conducted in non-premixed flames and therefore this review will be focusing on this type of flames, but its counterpart (premixed flame) will also be explained where needed. Non-premixed flames have been subject to intensive research and many books are available explaining their fundamentals. In this section, a brief summary of non-premixed flames is presented and this review was based on Turns [1], Warnatz et al. [2] and Renard et al. [4].

Flames can be divided into two configurations, namely premixed and non-premixed flames. Fuel and oxidiser are thoroughly premixed prior to combustion for premixed flames, while in the non-premixed configuration the fuel and oxidiser are mixed just before combustion mainly by molecular diffusion. Non-premixed systems are often called “diffusion flames” due to that reason. Many practical devices employ the non-premixed configuration over its counterpart due to safety reasons, as in premixed flames the possibility of sudden explosion as a result of premixing is rather high. A broader spectrum of stability also can be achieved in non-premixed and this is another key reason it is widely used, for example in diesel engines, gas turbines, furnaces etc.

Further, there are two types of non-premixed flames, namely normal diffusion flame (NDF) and inverted diffusion flame (IDF). In the normal diffusion flame, the fuel is flowed through the nozzle jet and oxidiser in the co-flow while in the inverted diffusion flame, this arrangement is swapped. While the application of the NDF in the real world has been mentioned in the previous paragraph, the IDF has extensively been used in many
laboratories, mainly as a tool to understand the structure and formation of soot, carbon monoxide and polycyclic aromatic hydrocarbon (PAH)\textsuperscript{2} such as in [12] and references therein.

Regardless of the arrangement, a non-premixed flame consists of separate fuel and oxidiser regions, hence the balance between the fuel and oxidiser normally characterised by the fuel-air equivalence ratio, $\phi$, as in premixed flame is rarely used. Instead, mixture fraction, $f$, is used to represent the degree of mixing and can be written as

$$f = \frac{Y_i - Y_{i2}}{Y_{i1} - Y_{i2}}$$

(2.1)

where $Y_i$ is the mass fraction of the element $i$, and the index ‘1’ refers to the oxidiser and index ‘2’ refers to the fuel. As the mixing proceeds, the mixture fraction, $f$ thus takes values

![Figure 2-1: Illustration of the appearance of flames. As the jet Reynolds number increases, laminar flames become longer, until they transition into a turbulence-dominated mode where flame length is no longer proportional to Re. Adapted from Warnatz et al [2]](image)

\textsuperscript{2} PAH, polycyclic aromatic hydrocarbons, are organic compounds that can be formed in incomplete combustion of organic materials such as coal, oil and petrol and are generally considered soot precursors.
between 1 and 0; at the pure fuel region \( f = 1 \), for a stoichiometric\(^3\) mixture, \( f = f_{st} \) and in the pure oxidiser region \( f = 0 \). The fuel rich region is the region where \( f > f_{st} \) and the fuel lean is the region where \( f < f_{st} \).

Non-premixed flames are further divided into three different classes which are known as laminar, transition and turbulent as shown in Figure 2-1 and this depends on the jet Reynolds number. In this thesis, the Reynolds number, \( Re \) is defined as the ratio of inertial forces to viscous forces and read as

\[
Re = \frac{\rho u D}{\mu}
\]  

(2.2)

where \( \rho \) is the density, \( u \) is the velocity, \( D \) is the diameter of tube and \( \mu \) is the dynamic viscosity of the fluids. Note that at lower Reynolds number the jet velocity plays an important role in the flame length, however once turbulence dominates (i.e. in the turbulent region where flow motion becomes chaotic), the proportionality of the flame length to the Reynolds number is no longer valid.

In the turbulent flames, there is a possibility (depending on the fuel/ oxidiser mixture) of domination of molecules diffusion over the turbulence in the near field region (below the transition point). At this region, the ‘local’ \( Re \) is reduced as a result of high dynamic viscosity generated by high local temperature. This effect, however, reduces as the \( Re \) is increased.

### 2.2.1 Laminar non-premixed flames

Laminar non-premixed flames have always been employed to understand flame structure and are used as a first step in modelling turbulent flames. Without the mixing caused by turbulence which complicates the flame, the laminar flame from the simulation can be easily compared side-by-side to the experimental measured data. This enables the influence of radiation, kinetic mechanism and diffusion model of a particular flame to be studied in detail.

The physical structure of the laminar flame has been described by Turns [1] as illustrated in Figure 2-2. Comparisons are made at three different axial positions, \( z = 0 \), \( z \) and \( z = L_f \). The

\(^3\) Stoichiometric mixture is the “perfect” mixture ratio of air to fuel; both fuel and oxygen would be completely consumed during combustion.
temperature (fuel temperature, $T_f$ and oxidiser temperature, $T_{ox}$) and mass fraction profiles (fuel mass fraction, $Y_f$, oxidiser mass fraction $Y_{ox}$, and product mass fraction, $Y_{pr}$) from the nozzle centre to the flame edge are presented in the left and right graph respectively. At $z = 0$, the mixing between the fuel and oxidiser is yet to take place. Further downstream at $z$, the mixing process through molecular diffusion is shown to diffuse the fuel outwards while the oxidiser is shown diffused inwards. In this example, the flame region is regarded as the stoichiometric region. The product fraction ($Y_{pr}$) is indeed at its maximum, close to where the peak temperature is achieved. At $z = L$, the fuel has all been consumed. This is a fuel lean region $f < f_{st}$ where the oxidiser exceeds the fuel fraction.

It can also be noted that from the nozzle exit, the flame expands and then narrows, creating a flame tip at the top. This happens due to buoyancy forces resulting from the generation of hot products which increase the local velocity and force the streamlines to get closer together. This increases diffusion due to the high gradient in the fuel fraction. Depending on the type of fuel used in the laminar non-premixed flame, the possibility of the soot formation is rather high, and linked to the fuel rich region. Note, however, that some soot

---

Figure 2-2: Illustration of laminar non-premixed flame. Adapted from Turns [1]. The left graphs are the temperature profiles, while on the right are the mass fraction profiles of fuel, oxidiser and products at different axial positions.
will be consumed when it enters regions plentiful in oxidiser. Soot formation has been studied extensively using laminar non-premixed flames employing fuels such as ethylene [13].

2.2.2 Turbulent non-premixed flames

Turbulent combustion is perhaps the most complex area in combustion research. The understanding of this area is far from sufficiently matured to apply a well-developed mathematical formulation as in the laminar flame. Turbulent combustion couples complex turbulent flow with additional variables such as heat release and chemical kinetics. Unsteady turbulent flow induces velocity fluctuations and affects all other combustion scalars such as the species concentration, temperature and density.

Considering Figure 2-3, a well-developed flow that is ejected from the fuel tube is perturbed at its boundary, and this generates a Kelvin-Helmholtz instability [14]. As it moves further downstream, the velocity difference between the jet and the co-flow increases, and this enhances the generation of the vortex. The surface area as well as the amplitude of these vortices is thus further increased with axial position leading to enhancement of the mixing process. A higher turbulent intensity can be achieved by increasing the jet velocity until the point when the mixing rate is so high that it competes with the chemical reaction rates, creating the possibility of so-called “localised extinction”, where the chemical reaction stalls

![Figure 2-3: Illustration of vortex generation in turbulent non-premixed flame. Adapted from Renard [4].](image-url)
Further gradual increase of the jet velocity would then un-anchor the flame from the jet tube rim but it will be stabilised at a point downstream to create a “lift-off” or “lifted” flame. The distance from the stabilisation point to the fuel tube rim is called the lift-off height. In this scenario, the space between the rim and the flame enables mixing between the fuel and the oxidiser, so this zone contains both non-premixed and premixed gases. Watson et al. [16] undertook a study to understand the cause which triggers a lifted flame and how it is stabilised in the case of a diffusion jet flame. They suggested that localised extinction happens due to radial movement of a fuel pocket or oxidiser pocket (imaged using Rayleigh⁴) into the reaction zone layer (imaged using CH-LIF (laser-induced fluorescence)), which reduces temperature by means of enhancing heat conduction out of the reaction zone into the fuel rich or fuel lean regions [17]. The flame is stabilised by the existence of hot products downstream which keep re-igniting the incoming premixed fuel. As velocity is further increased and depending on the degree of the premixing, global extinction will eventually occur.

The localised extinction phenomenon in highly turbulent flames is really challenging as a reliable model needs to accurately capture the finite-rate chemistry effect from such events. To understand the response of species mass fraction towards the turbulence as well as localised extinction, Barlow and Frank [18] performed a simultaneous Rayleigh/Raman and CO-TLIF (two-photon laser-induced fluorescence) experiment in a partially premixed piloted jet. They reported that the decrease of Damköhler number, $Da$ and increase of axial position would affect the mass fraction of the main species. The Damköhler number, $Da$ is defined as

$$Da = \frac{\tau_m}{\tau_c}$$

(2.3)

where $\tau_m$ is the mixing rate and $\tau_c$ is the chemical reaction rate. A numerical model without considering this effect (species mass fraction shifting due to local extinction) would not represent an accurate result.

A complete understanding of the interaction between the flame chemistry and turbulent mixing should also cover knowledge of the dissipation structures of the flame itself. A

⁴ Please note that the laser techniques will be explained in detail in Chapter 3.
turbulent flame consists of a wide range of length scales, but in the past only the largest was resolved by experiments. Recently, the smallest scale of fluctuations – which is known as the Batchelor scale, $\lambda_B$ – has been successfully resolved by Wang et al. [19] in temperature and mixture fraction measurements using Rayleigh/ Raman and TLIF-CO techniques. The high SNR of one-dimensional temperature images were used to obtain one-dimensional energy spectrum. A cut-off wavelength, $\lambda_c$ at 2 % of the peak was then determined from the spectrum and further $\lambda_B$ can be calculated using $\lambda_B = \lambda_c / 2\pi$. In another study, Frank and Kaiser [20] demonstrated two-dimensional temperature derived from high-resolution Rayleigh signals were also able to resolve thermal thickness layers. This information is beneficial for example in understanding the relationship between the jet Reynolds number and heat release. This information could also be used to further improve the numerical model to accurately predict the combustion process. The thermal thickness analysis will be discussed in greater detail in Chapter 7.

2.2.3 Inverted flames

The Inverted Diffusion Flame (IDF) configuration has historically been used to study soot formation. The soot formation was reported by Shaddix et al. [21] to be influenced by several factors: 1) the overlap of high temperature and fuel rich or fuel lean regions, 2) the residence time of soot in low temperature regions, and 3) the thermophoretic\(^5\) effect. In IDF, the overlap of the high temperature and the fuel rich regions is thinner compared to the one in NDF [21]. At this region, the NDF indeed produces higher probability of soot formation compared to the IDF. However, this is compensated by the steeper temperature gradient in the fuel lean region in IDF configuration which transports away the soot into the fuel rich region by the thermophoretic effect that eventually enhances the soot growth. This will be further enhanced by the lower vertical velocity in fuel rich region which enable longer residence time in that fuel rich region. By contrast, soot in the NDF will be transported downstream to the high temperature oxidation zone and get oxidised. However, the thermophoretic effect and longer residence time are not enough to generate higher soot than the NDF [21].

\(^5\) The thermophoretic or Soret effect is the response of a particle/molecule towards the force of temperature gradient. Positive Soret implies the particle/molecule moves away from hot towards cold region while negative Soret implies the particle/molecule moves closer to hot region.
An extensive study has been made by Wu and Essenhigh [22] where they mapped six different types of methane IDF based on appearance and stability. Temperature profiles together with stable species of IDF and NDF measured experimentally were also compared to numerical predictions. Subsequently, Takagi et al. [23] investigated a H$_2$ diluted N$_2$ flame and found the IDF produces higher flame temperature near the flame tip compared to NDF resulting from higher H$_2$ ratio and excess enthalpy accumulated in the central part of the flame.

Lee et al. [24] observed the level of partial premixing increases with increasing level of inner tube recess as well as velocity ratio ($V_{air}/V_{fuel}$). The flame height can be reduced by a small increase of air flow rate. Flame stability and lift-off height were also examined and reported. Sobiesiak and Wenzell [25] extended their study by varying nozzle and velocity ratio. They reported the presence of partial premixing in the non-premixed inverse flame following the analysis of temperature profiles and Schlieren images. They also concluded that velocity ratio has an impact on the flame length. Proper optimisation of velocity ratio, fuel/air nozzle diameter ratio and equivalence ratio could also produce an extended region of uniform and high temperature.

Other types of IDF burner configurations were studied by Sze et al. [26]. They investigated flame appearance and temperature distribution in inverse circumferentially arranged ports (CAPs) and co-axial (CoA) jets. Flame length was observed to decrease with increase in airflow rate at fixed fuel velocity. This is due to stronger entrainment between the fuel and the air through premixed mode. With high enough airflow rate, the premixing was reported to further enhanced enabling all the fuel to be burned. They also reported with increasing jet velocity, that the flames become bluish from yellowish resulting from non-premixed to premixed combustion, and CAP mixing rate is more intense so transition occurs at lower velocity compared to CoA. The temperature was also reported to be higher in the CAP than the CoA flame. Another recent study performed by Elbaz et al. [27] used high speed OH-PLIF and 2D PIV measurement to temporally understand the mixing and entrainment in an IDF by varying the velocity ratio in a methane flame. They also concluded that increasing the velocity ratio enhances the fuel-air mixing through increased entrainment of the fuel towards the air jet which supports the findings of earlier measurements [24-26].
2.3 Role of laser diagnostics in combustion

Laser diagnostics have helped the combustion community to understand combustion phenomena in great detail. Continuing effort has been applied to keep improving the capability of laser diagnostics to improve the measurement, especially on the spatial and temporal resolution as well as to improve the accuracy. For example, in the past thermocouples were used to measure the temperature, however the radiative correction that was needed would introduce a large error in the temperature reading. The need of the probe to be in the flow field also becomes a problem as this clearly would perturb the flow. Combustion involves complex chemical reactions, and intermediate species such as hydroxyl (OH) and formaldehyde (CH\textsubscript{2}O) cannot be detected by a gas sampling sensor. These species however are easily detected using laser diagnostics techniques.

Other than the non-intrusive capability, high spatial resolution is also offered, and a resolution of 50 μm is easily achieved. Depending on the measurement of interest and the area of imaging required, a higher spatial resolution at around 20 μm is also possible as shown by Frank et al. [20, 28] in their turbulent structure investigation. Good temporal resolution using a repetition rate of 10 Hz is also easily achieved. High speed lasers (several-kHz) repetition rate in the past only comes with a low energy laser which might be enough for certain measurement such as for laser induced fluorescence (LIF) technique. However this limitation has been improved with high-energy pulse burst laser systems (HEPBLs) as reported by Papageorge et al. [29] that can produce a high-repetition, high-energy laser (for a finite number of shots). This development has enabled kHz rate Rayleigh scattering, a technique which requires a high laser energy to achieve a high signal to noise ratio (SNR). Another important aspect when dealing with turbulent flow is the capability to ‘freeze’ the flow evolution during sampling. A very short pulse length, at around 10 ns, can be achieved using laser diagnostics.

There are several measurement techniques available to be used in combustion studies. Depending on the measurement of interest (temperature, velocity etc.) and the requirements (SNR, spatial and temporal resolutions etc.), a specific technique or even joint –techniques (multi-scalar measurement) could be performed. This current work involves the measurement of species concentration (OH and CH\textsubscript{2}O – PLIF) and temperature. A review of
the laser diagnostic techniques used in the literature for these types of measurements will be discussed next.

2.3.1 Temperature measurement

The highest accuracy in temperature measurement is often said to be achieved using the coherent anti-Stokes Raman scattering (CARS) technique. CARS probes the same Raman active mode as in the Raman scattering. However, in CARS a nonlinear wave-mixing process is used to enhance the weak Raman signal. CARS requires two laser beams, known as pump at wavelength $\omega_p$ and Stokes at wavelength $\omega_s$ to interact with the species. A strong anti-Stokes signal at $\omega_{as} = 2\omega_p - \omega_s$ is generated as a result of coherently driven molecular oscillation when the beat frequency $\omega_p - \omega_s$ matches the frequency of a Raman active molecular vibration. In the past, temperature measurement has largely been performed using nitrogen or hydrogen CARS. The reliability of using these two different molecules has been investigated by Hancock et al. [30] and Hussong et al. [31] and references therein. The average temperatures measured using hydrogen CARS were reported to be higher than nitrogen CARS under the same conditions and higher even than calculated equilibrium temperatures which were thought to be due to the linewidths and line-broadening mechanism of the hydrogen itself. This temperature deviation mainly occurs in the fuel rich region. The improvement of CARS in terms of expanding to other molecules to be the temperature probe such as CO$_2$ [32, 33], H$_2$O [34, 35] and O$_2$ [36] has always been of interest. A further improvement of CARS has been made recently to measure temperature in an oxy-fuel burner furnace using O$_2$ CARS which was capable of measuring a temperature in the range between 400 K – 3000 K [37]. The accuracy of that specific system was not mentioned, however O$_2$ dissociation occurring at high temperature was stated to be around 11 % which in turn resulted in a reduced accuracy. Applicability of CARS in harsh environments is another advantage of CARS, however CARS also suffers from the so-called “spatial averaging” problem which was reviewed by Parameswaran et al. [38]. This problem is mainly attributed to spatially varying temperature and composition which then leads to a bias in the estimated flame temperature. The other limitation is that it can generally only be used as a point measurement. 2D-CARS was recently demonstrated by Kliewer and Bohlin [39] however its applicability to flame measurements is not yet fully established. In respect
of the accuracy of CARS, Miller et al. [40] reported 2.6 % shot-to-shot variations with 2.6 % of error for temperature measurement performed at room temperature. In O$_2$ heated jet, the error was reported to be 9.4 %.

Another technique that makes temperature measurement possible is laser induced fluorescence (LIF). LIF is a species specific technique, where a laser beam is tuned to excite a specific transition of an interest species. The incident light is absorbed by a fraction of molecules at the ground state and these molecules will then be promoted to a higher energy state. After a very short time, a fraction of the excited molecules re-emit the energy and fluorescence is one of several processes that takes place for the molecules to come back to its original state. Other processes include collisional quenching, predissociation, photoionisation and stimulated emission. The last three processes mentioned here are negligible, but collisional quenching significantly competes with fluorescence. An accurate temperature-inferred measurement from OH-LIF hence requires knowledge of the mole fraction of the species involved so a collisional quenching correction can be calculated, which is very challenging in a turbulent flame. This could however be avoided by employing two line laser induced fluorescence (TLIF) as the temperature is determined from the ratio of signals coming from two temperature-dependent transitions [41]. TLIF is often used in sooting environments as well as in combustion engines where Rayleigh scattering thermometry is impossible. The choice of the excited molecule or atom depends on the existing molecules in that particular region, or else seeder molecules could be used. For example, Tamura et al. [42] seeded nitric oxide (NO) in their flame, however consumption of this molecules was observed as NO is an active species which reacts with CH producing HCN and O. Another investigation performed by Nygren et al. [43] where a simultaneous measurement of Indium atoms (seeded) and OH from the flame were used in a sooting environment has shown that OH thermometry works best in higher temperature regions, while the indium atoms are effective in the rich or colder parts. Thus, a clear strategy has to be decided before a thermometry measurement could be performed. The main limitation of this approach is the temperature range which is linked to the excited species: OH is only in the post flame region, while indium is reported to being sensitive in a range of 700 – 3000 K [44]. However due to the high signal levels produced by the fluorescence of the molecules or atoms, a two-dimensional measurement is possible.
Another optical technique often used for temperature measurement is Rayleigh scattering. This technique is popular as it has a strong signal (up to $10^3$ greater than Raman), enabling two-dimensional imaging. Rayleigh scattering is an elastic scattering process where the elastics collisions between gas molecules and incident light scattered the photons at the same wavelength of the laser. The intensity of Rayleigh scattered light is proportional to the laser power, number of density of species involved and Rayleigh cross-section. In a constant pressure situation, and if the Rayleigh cross-section is constant throughout the combustion process (reactants, intermediates and products), the temperature can be directly inferred as it is inversely proportional to the Rayleigh signal. The applicability of this technique was shown by Dibble et al. [45] and Bergmann et al. [46]. This however requires a carefully tailored fuel and oxidiser mixture. ‘Traditional’ Rayleigh scattering (‘traditional’ Rayleigh will be used to represent a Rayleigh technique considering constant Rayleigh cross-section hereafter) can only be applied in a non-sooting environment, and suffers with contamination from scattered light from surfaces and particles as the wavelength of the collected scattered light from the gas molecules is the same as the incoming electromagnetic radiation. This unwanted scattered light however can be eliminated by employing a filtered Rayleigh scattering (FRS) technique which was introduced by Miles et al. [47]. There are several filter candidates for FRS that have been reported such as mercury [48] and iodine [49] which were employed using a tripled Alexandrite laser at 254 nm and frequency doubled Nd:YAG laser at 532 nm respectively. The working principle of FRS is the same as ‘traditional’ Rayleigh scattering, other than adding a molecular filter cell in between the light collection device and the flow field to filter the light. In FRS, the narrow absorption line filter is used to block Mie scattering which is produced by the stationary or slowly moving particles and thus essentially not broadened spectrally while transmitting the broadened molecular Rayleigh scattering which results from the Doppler and collision broadening processes acting on the molecules.

In order to measure temperature when using more complicated mixtures such as found in oxy-fuel flames, ‘traditional’ Rayleigh (a constant Rayleigh cross-section) cannot be used. Another variant measuring both polarised and depolarised Rayleigh scattering signals takes advantage of isotropic and non-isotropic species to determine some information about the mixture composition and thus the relative Rayleigh cross-section and ultimately recover the
temperature. Fielding et al. [50] and Frank et al. [51] have successfully shown the applicability of this technique and it will be used in this work to measure oxy-fuel flame temperature. Further discussion is presented in section 3.1.2 and Chapter 5.

2.3.2 Concentration measurement

Raman scattering is very popular in combustion studies since it can characterise the species involved in the combustion process. Unlike Rayleigh scattering where the scattering molecules return to the original state and thus the re-emitted photon is at the same wavelength of the incoming electromagnetic radiation (elastic process), Raman is species specific as the molecules do not return to their original state (inelastic process), rather to a higher energy state than the original (Stokes) or lower (anti-Stokes) as shown in Figure 2-4. This difference between the absorbed and re-emitted energy is equal to the vibrational energy spacing, and every molecule has different vibrational energy level spacing. The small energy difference in turn produces a different emission wavelength and enables the detection of different species. Raman scattering however suffers from a weak signal due to a small Raman cross section and hence only point and line measurements are possible. Raman scattering enables simultaneous detection of several species such as H₂, H₂O, CH₄, CO, O₂ and N₂ [52] and this technique is considered well developed for CH₄-air [53, 54], H₂-
air[55] and CH₄/H₂/N₂-air [46, 56] flames. The Raman technique has also been applied in oxy-fuel flame studies [57, 58].

For other intermediates such as OH, CH₂O, CH and NO that cannot be detected by Raman, a single photon LIF technique is frequently applied. As explained in section 2.3.1, the LIF signal is relatively high which makes two-dimensional imaging possible. Other possible species that could be measured include O [59] and CO [60] using TPLIF (two-photon laser-induced fluorescence). The limitation, as explained in section 2.3.1, is that data collected using this technique is only qualitative if collisional quenching is not corrected, for example using the data provided by Tamura et al. [61]. One key intermediate, CH₂O, cannot be fully quantified due to insufficient quenching data. One approach to yield moderate accuracy is to use a power law correction assumption [62]. The quenching correction will be explained further in sections 3.1.3 and 5.2.2.

2.3.3 Multi-scalar measurements

Experimental data is often used to validate numerical modelling results. Numerical calculations produce a large range of outputs which include all the major species and most of the minor species in a combustion process. Using only one quantity, for example a comparison between the measured and calculated temperature would not be enough to conclude the numerical model is reliable. More sophisticated comparison by means of validating several quantities is therefore required. This could be achieved by performing a simultaneous multi-scalar measurement, where two or more quantities are collected at the same time.

A simultaneous single point measurement is often sought, which can be performed using simultaneous Raman-Rayleigh or Raman-Rayleigh-LIF measurements to collect as much information as possible towards the initial understanding of the specific flame. These types of investigations have been performed by Sevault et al. [57] in air-like temperature (flame temperature below 2200 K) turbulent non-premixed oxy-fuel flames and Fuest et al. [63] in piloted jet dimethyl ether laminar and turbulent flames employing the former and latter technique respectively. Raman scattering is usually performed simultaneously with the Rayleigh measurement, as the temperature can be deduced accurately using the known species concentration measured in Raman scattering [57]. In the dimethyl ether
investigation performed by Fuest et al. [63] however, the species from Raman scattering measurement alone are not enough to accurately determine the temperature. This is because Raman scattering only able to detect major species while other minor hydrocarbon species that are negligible in CH₄-air flame contributes for about 16 % in the DME-air flame. If these minor species are not considered, an unacceptably large systematic error is introduced. Hence the CO-LIF measurement and numerical simulation data are required to accurately infer the temperature.

The point measurement discussed earlier is not enough to understand the evolution of the combustion process. Simultaneous, two-dimensional imaging should be performed as this generates more understanding through the visualisation of the single-shot images. A simultaneous velocity measurement by means of PIV and reaction zone identification by means of OH-PLIF has been performed in turbulent premixed flames to characterise the turbulent flux of mean reaction progress variable [64]. Using a similar joint-measurement technique, Rehm et al. [65] investigated the correlation between strain rates, vorticity and reaction zone structure in laminar and turbulent non-premixed jet flames. Based on the same principle, Carter et al. [66] investigated a relationship between the flame front layer using CH-PLIF and flame front strain rate in a non-premixed CH₄-air flame. The velocity and OH-PLIF joint measurement is relatively simple in laser diagnostics studies. It is also beneficial as lots of data could be derived from the velocity-flamefront images. Since these species (OH and CH) only exist in a small portion of the overall flame, a further understanding of the overall flame structure is required. Most et al. [67] have successfully investigated the correlation between the temperature and velocity where single shot images were able to generate information about the curvature and temperature gradients (from temperature image) and vortices, flow divergences and strain rates (from velocity image).

A significant effort has also been put to measure flame heat release. Najm et al. [68] reported that heat release is spatially and temporally correlated to production of HCO. However, HCO concentration is very small and thus HCO-PLIF imaging is impossible [62, 68]. Paul et al. [62] then showed that HCO correlates with multiplication of OH and CH₂O LIF signals taken at the same spatial location, in other words that heat release is proportional to the product [OH][CH₂O]. A number of investigations have been performed to understand
the heat release in premixed [69, 70] and in non-premixed [71, 72] flames by means of joint-measurement of OH and CH$_2$O PLIF. Gordon et al. [71] and Medwell et al. [73] performed simultaneous temperature (using ‘traditional’ Rayleigh technique), OH and CH$_2$O PLIF experiment in their study, however the Rayleigh scattering cross-section variation is rather high, at more than 20 % (the temperature is also expected to have this much error) which negatively affects the accuracy of the quenching correction made to the OH and CH$_2$O signals. A combination with the polarisation/ depolarisation Rayleigh technique reported by Frank et al. [50, 51, 74] would be an effective way to reduce the uncertainty in the temperature measurement.

2.4 Oxy-fuel flames overview

2.4.1 Differential diffusion effect

Differential diffusion has been a major subject in combustion mainly in numerical modelling. Despite this, it has not always been included in models. Instead, it is often assumed that all species diffuses at the same rate to simplify the numerical model, the so-called “equal diffusivity transport model”. However, it has been shown by many investigators that differential diffusion is important especially near the jet nozzle – for example, by Bergmann et al. [46] and Meier et al. [56] which suggests the multi-component transport model, where the diffusion of all species are considered, needs to be used. Both of these investigations were performed using CH$_4$/H$_2$/N$_2$ - air flames. The measured main species and temperature data were compared to several set of calculated data (calculated at different strain$^6$ rates and Lewis$^7$ number). Meier et al [56] concluded that the thermochemical state of the flame is complex; at the upstream region, a single strain rate and Lewis number would not be able to perfectly mimic the measured data (radially, from the nozzle centre to the edge of the flame). The strain rate changes along the radial location, with higher value at the jet centre and decreases near the shear layer. This is due to the reduced velocity (near the shear layer) as a result of flow mixing between the fuel and oxidiser. The molecular diffusion also becomes less important near the shear layer region. The behaviour of strain rates and Lewis

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$^6$ Strain rate quantifies the speed at which the flame departs from chemical equilibrium and has units of s$^{-1}$

$^7$ Lewis number (Le) is the ratio of thermal diffusivity to mass diffusivity.
number for the flames investigated in this thesis will be explained in greater detail in section 5.3.4.

In the oxy-fuel flame specifically, Sevault et al. [57] compare the experimental temperature data to the calculated temperature at different strain rates and Lewis numbers. As expected, for locations near the jet exit, a numerical calculation that considers a higher strain rate, \(a=800s^{-1}\) and multi-component diffusivity considerably better matches the experimental data. Strain rate was shown to decrease to \(a=15s^{-1}\) further downstream and the diffusion model switches to equal diffusivity near stoichiometric conditions, but multi-component diffusion is important in the fuel rich and fuel lean regions.

Recently, the effect of differential diffusion in an oxygen enhanced counter-diffusion methane flame has been studied numerically by Dietzsch et al. [75] and the result was compared to a normal CH\(_4\)-air flame in laminar and turbulent environment by assessing the C and H atoms’ mass fractions. In a laminar CH\(_4\)-air flame, the atoms show no sensitivity to differential diffusion. The same result was shown for the C atom in an oxygen-enhanced laminar flame, however H atoms were highly affected by differential diffusion. By contrast, the H atom was less affected by differential diffusion in a turbulent oxygen-enhanced flame, which was reported to be due to the flow characteristics being dominated by turbulence.

### 2.4.2 Soot formation and flame radiation

A considerable number of studies have been performed to investigate soot formation and flame radiation in oxygen-enriched (oxidiser mixture of N\(_2\) and O\(_2\)) and oxy-fuel (oxidiser mixture of O\(_2\) and CO\(_2\)) combustion. Maximised radiative heat transfer is desired in oxy-fuel combustion applications as this enables a uniform temperature distribution through the high concentrations of CO\(_2\) and H\(_2\)O [76, 77]. The existence of soot in the flame has also been reported by Liu et al. [76] in contributing to the enhancement of radiative heat transfer, however from the laser diagnostics point of view, especially when employing the Rayleigh scattering technique, this should be avoided.

In natural gas combustion, as expected Baukal et al. [78] observed thermal radiation to increase when increasing the oxygen content from 28 % (the remainder is N\(_2\)) to 100 %. Using the same principle of varying the quantity of O\(_2\), Wang et al. [79] expended the
experiment to propane and blended fuels as well as by varying the jet Reynolds number from Re=5000 to Re=15000 and comparing it with a natural gas flame. Increasing O₂ was shown to moderately increase the radiant emission, with the propane fuel always higher than the blend and natural gas. The increase of Reynolds number from Re=5000 to Re=15000 was also reported to affect the radiant fraction, decreasing it by 33 %, as the probability of soot formation is inversely proportional to the Reynolds number. Soot formation however was shown to increase with increasing O₂ concentration, to a peak at 40 % and reducing with further addition of O₂.

In the latest study performed by Shaddix and Williams [80] using a methane oxygen enriched flame, the same behaviour is observed in soot formation and thermal radiation as reported by Wang et al. Soot formation was shown at a minimum level below 100 mm height above burner and was insensitive to variation of O₂ or Reynolds number. In another study by Beltrame et al [81] which related soot loading with the variation of strain rate between 10 – 60 s⁻¹, it was found that soot quantity decreases with increase in the strain rate. It was reported that the reason was due to the short residence time at high temperature to develop the soot. To date, no investigation on the soot formation in O₂/CO₂ mixtures has been reported, though an investigation by Ditaranto and Oppelt [82] of thermal radiation showed an increase with increasing O₂ concentration, however this is not enough to conclude the soot is generated in the same pattern as it might only come from H₂O and CO₂, as reported by Shaddix et. al. [80].

2.4.3 Flame chemistry

Significant effort has also been put towards understanding the chemistry side of oxy-fuel flames. As oxygen-enhanced oxy-fuel flame applications use a high concentration of O₂ (> 40 %), this will in turn produce a high temperature flame (> 2200 K). Thus, thermal dissociation mainly of the CO₂ molecules, production of NOₓ, and production of radical pools (O, H and OH) have to be taken into account when modelling an accurate oxy-fuel flame reaction mechanism to compare with experiments, all of which will now be discussed.

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8 Radiant fraction is defined as the ratio of radiative heat loss to the total heat release
CO production from CO2 has been widely accepted to occur in oxy-fuel flames [83, 84] (and references therein); the CO2 is no longer inert and instead participates in the chemical reaction at high temperature [83, 85, 86]. This results in an increase of CO in oxy-fuel combustion compared to air fed combustion, as reported by Andersson et al. [87, 88] with propane fuel. To make a fair comparison, the oxygen in the oxy-fuel flame was adjusted so both flames produce the same temperature. The higher CO production in oxy-fuel was reported to possibly come from the dissociation of the CO2 and/or the reaction of this stable species with other intermediates.

Extensive investigations have been performed to understand this reaction especially in turbulent flames, through modelling as well as experiments. In diffusion modelling, Liu et al. [83] investigated the effect of adding the CO2 in the fuel and in the oxidiser. CO2 added into the oxidiser was reported to produce significantly higher CO concentration than the same amount added into the fuel stream. In premixed methane flames, an investigation performed by Glarborg et al. [84] covers both modelling and experiment using N2 and CO2 as inerts, with the maximum temperature of about 1800 K. The modelling result was compared to the experimental result, and further the key reactions involving CO2 were identified. As expected, the analysis shows CO is mostly generated by the reverse reaction of CO2 and H, 

\[ \text{CO} + \text{OH} \leftrightarrow \text{CO}_2 + \text{H} \]

at medium to high temperature. CO2 was also shown to react with methylene, CH2 which this accounts for about 20% of production in these types of flames. At high temperatures of around 1700 K – 1800 K, the dissociation of CO2 to CO was reported to start taking place. Recently, Chen and Ghoniem [8] performed thermodynamic analysis on the numerical result calculated using CHEMKIN employing the GRI-Mech 3.0 detailed mechanism, and realised that other than CO, more H2O is also produced in the oxy-fuel flame compared to the air flame, which was reported as a result of interaction between OH and H2, 

\[ \text{OH} + \text{H}_2 \leftrightarrow \text{H} + \text{H}_2\text{O} \]

The idea to replace N2 with CO2 in oxy-fuel combustion is to suppress or even remove the production of NOx and to enable condensation separation techniques for CO2 capture. However, it was reported by Sung et al. [89] that there is still a small fraction of NOx generated and this comes from leakage into the oxidiser or from the fuel source. The production is indeed far smaller than air-fuel flame combustion [83, 90].
The other concern regards the radical pool (O, H, and OH) that exists in oxy-fuel combustion. A numerical study performed by Seepana and Jayanti [91] shows the radical pool in an oxy-fuel flame is smaller compared to the air flame. The calculation is, however, based on a rather low temperature of about 2100 K. Another study that discussed this radical pool was done by Linow et al. [58] in an H₂/O₂ flame showed that the radical pool totals about 10% of the overall volume fraction. This shows that for high temperature oxy-fuel, the experimentalist has to take this into consideration. The presence of these radicals has always been neglected in air flame combustion.

2.4.4 Flame speed and stability

An investigation of flame speed is important especially in determining the stability of a flame to prevent events such as blow off or flashback. As the N₂ is replaced by CO₂ in oxy-fuel combustion, the burning velocity is also affected due to the change of transport, thermal and chemical reaction mechanisms. High thermal radiation from CO₂ also contributes to the change of flame speed [85]. Relevant investigations of the flame speed in oxy-fuel combustion are now discussed.

Liu et al. [85] performed a numerical study on the effect of replacing N₂ with CO₂ in CH₄ and H₂ premixed flames. The result was compared to H₂-air and CH₄-air premixed flames. The replacement with CO₂ was reported to significantly reduce the burning velocity as compared to both H₂-air and CH₄-air. Again, this reduction was reported due to the reverse reaction of \( CO + OH \rightleftharpoons CO₂ + H \) where CO₂ competes with O₂ for chain branching reaction, \( H + O₂ \rightleftharpoons O + OH \) which in turn reduces the presence of important radicals (O, H, and OH) that support the flame speed [85].

In a study by Benedetto [92] using CH₄/O₂/N₂/CO₂ and H₂/O₂/N₂/CO₂ mixtures in closed cylindrical vessels, a reduction in burning velocity was observed with increase of CO₂. However, the reduction of burning velocity was reportedly due to the lower flame temperature as a result of reduced combustion rate. It was argued that added CO₂ increases the specific heat, in contradiction to Liu et al. [85] who reported the lower burning velocity is the result of a chemical kinetics effect. Chan et al. [93] then later reported that the kinetics effect only takes place at a higher CO₂ concentrations.
In regards to the flame stability, it was reported that if N\textsubscript{2} is replaced with CO\textsubscript{2}, more than 21 % of O\textsubscript{2} is needed to achieve an air-like flame \cite{94}. The reported reason was due to the lower laminar burning velocity as CO\textsubscript{2} has higher density and lower thermal conductivity. Furthermore, high concentration of CO\textsubscript{2} and H\textsubscript{2}O in an oxy-fuel flame reduces the flame temperature as these molecules have high molar heat capacity compared to N\textsubscript{2}. Other than the reactants, the stability also depends on factors such as the type of burner, Reynolds number of the nozzle jet, as well as reactants involved in the combustion.
3 Experimental techniques and equipment

3.1 Background on measurement techniques

3.1.1 Hot wire anemometry

The characterisation of velocity profile and boundary condition of the burner used in this research was performed using one dimensional hot wire anemometry (HWA) probe in conjunction with a constant temperature anemometer (CTA) module. Due to the maturity of this HWA technique, only technical aspects and the specifications of the system are described here and readers could find the working principles of this technique in great detail such as in Hoffmeister et al. [95].

A single platinum-plated tungsten of 1.25 mm length with 5 µm diameter wire sensor probe (DANTEC 55P11) was used as this type of wire has a high frequency response. The CTA system of DANTEC Measurement Technology, US comes with a built-in signal conditioning and A/D board units enabling electronic noise to be removed as well as preventing signal aliasing. A square wave test of the combination of the sensor and anemometer was performed, resulting a cut-off frequency of 22 kHZ at the standard -3 dB limit. A temperature probe was also connected to the bridge to account the temperature compensation of the measurement. Calibrations which establish the relationship between the velocity and output voltage of the probe were done before and after the measurement was taken, involving a 20-point velocity calibration which was then approximated through a 4th order polynomial curve fitting. Specifically for this experiment, based on the intended bulk flow velocities, a calibration range of 0.5 m/s – 80 m/s was carried out using DANTEC calibrator unit. For the velocity measurement, the probe was mounted to a three-axis precision stage, enabling the probe to be repositioned. The analogue signals were first filtered, and then sampled by a 16 bit A/D adaptor card at a sampling frequency of 20 kHZ, recording $2^{21}$ data points which were then stored using a LABVIEW in-house program. Please note, however the collected data from this HWA measurement was only used to get the mean and RMS velocity profile of the isothermal flow field.
3.1.2 Rayleigh scattering

The elastic scattering of light from atoms, molecules or particles with diameters much smaller than the light wavelength ($d < \lambda$) [96] is called Rayleigh scattering. As the photons are scattered without energy exchange, the frequency of the scattered light remains unchanged. Unlike Raman scattering where the Raman shift is depending on the structure of species, and thus concentration of particular species can be determined, this is not possible in Rayleigh scattering. However the Rayleigh signals can be used to determine the molecular number density, and hence the temperature in constant pressure situations can be resolved. This technique is very useful in turbulent combustion studies as it produces higher signal compared to Raman scattering, due to the fact that Rayleigh cross-sections are three orders of magnitude larger than vibrational Raman [97]. Laser Rayleigh scattering (LRS) however suffers from stray light mainly from Mie scattering (from particles which are larger than the light wavelength). This can be prevented by covering the laser beam and placing matte black wall while the Mie scattering can be avoided by running an experiment in a clean environment, free from any particles and dust.

The Rayleigh signal intensity is proportional to the effective Rayleigh cross-section, $\sigma_{Ray, eff}$. The latter is a summation of species-specific Rayleigh cross-section, $\sigma_{Ray,i}$ weighed by the mole fraction, $X_i$ of constituent $i$ which is expressed using

$$\sigma_{Ray, eff} = \sum_i X_i \sigma_{Ray,i} \quad \text{(3.1)}$$

In this work the Rayleigh cross-section of every species was taken from Fuest et al. [63] where they were derived based on the refractive indices of excitation wavelength at 532 nm taken from Gardiner et al. [98]. The Rayleigh cross-section calculation was reviewed by Miles et al. [99] suggesting the Rayleigh signals are contributed from three components namely Placzek trace scattering, Q-branch rotational Raman scattering, and O- and S-branches rotational Raman scattering. Taking into consideration these three components, for the linearly polarised incident light, the Rayleigh cross-section of each species, $\sigma_{Ray,i}$ in terms of depolarisation ratio, $\rho$ can be expressed as

$$\sigma_{Ray,i} = \frac{4\pi^2 (n_i - 1)^2}{N_0^2 \lambda^4} \left( \frac{6 + 6\rho}{6 - 8\rho} \right) \quad \text{(3.2)}$$
Where \( n_i \) is the refractive index, \( N_0 \) and \( \lambda \) are the number of density at standard temperature and pressure, STP \((2.69 \times 10^{25} \text{ m}^{-3})\) and laser wavelength respectively. This is true for all species, except for the \( \text{H}_2 \) where the pure rotational Raman lines are spectrally separated from the Rayleigh line when a narrow band filter is used. For the \( \text{H}_2 \), the expression becomes

\[
\sigma_{\text{Ray,}i} = \frac{4\pi^2(n_i - 1)^2}{N_0^2 \lambda^4} \left( \frac{6 + 6\rho}{6 - 8\rho} \right) \left( \frac{4 - 3\rho}{4 + 4\rho} \right)
\]  
(3.3)

The laser Rayleigh scattering signal, \( S_R \) (e.g. measured in counts as digitised by a camera) can be then expressed as

\[
S_R = C \cdot I \cdot N \cdot \Omega \cdot A \cdot l \cdot \sigma_{\text{Ray,eff}}
\]  
(3.4)

Where \( C \) is a calibration constant, \( I \) is the incident laser intensity, \( N \) is the number density, \( \Omega \) is the solid angle of signal collection from the measurement area, \( A \) and \( l \) are the focal area and axial length where the Rayleigh signal is observed, respectively. For a low velocity jet, the change of static pressure is neglected and hence the ideal gas equation can be used and substituted in the equation (3.4) gives

\[
S_R = C \cdot I \cdot \frac{P A_0}{RT} \frac{\Omega}{4\pi} A l \sigma_{\text{Ray,eff}}
\]  
(3.5)

Where \( P \) is the pressure \( A_0 \) is the Avogadro’s number, \( R \) and \( T \) are universal gas constant and temperature respectively. The absolute temperature, \( T \) is hence inversely proportional to Rayleigh scattering intensity.

From the equation (3.5), it shows that the temperature can be easily inferred from the Rayleigh signal providing the \( \sigma_{\text{Ray,eff}} \) is known. They are two ways of achieving this; 1) using a fuel/ oxidiser mixture that has a constant Rayleigh cross-section throughout the combustion process (from reactants, intermediates to product). This approach was adopted in fuel-air combustion by Dibble et al. [45]. In their study, 38 % methane (\( \text{CH}_4 \)) and 62% hydrogen (\( \text{H}_2 \)) by volume were used as the fuel which gives +/-5 % change to the cross-section, and by Bergmann [46], which using 22.1 % \( \text{CH}_4 \), 33.2 % \( \text{H}_2 \) and 44.7 % \( \text{N}_2 \) (also known as DLR-A) in the fuel stream resulting of +/-3 % cross-section variation from reactants
to products. 2) Determining the mole fraction information of each species as a function of location using either a combination of experiment and numerical model, as performed by Smith et al. [100], or simultaneous Rayleigh/ Raman experiment to deduce the species information form Raman scattering data.

In high temperature oxy-fuel flame, a constant Rayleigh cross-section is impossible to achieve. This is mainly due to the existence of high concentration of CO$_2$ that has a large Rayleigh cross-section and also CO$_2$ decomposition in high temperature region. The determination of species mole fraction from numerical model also is somewhat complicated in this complex fuel/ oxidiser mixture. A simultaneous Rayleigh/ Raman experiment in oxy-fuel flame to determine temperature and main species has been shown to work by Sevault et al. [57], however the flame temperature was around air-flame temperature. At this temperature, the intermediate species such as O, OH and H are at the minimum level. These intermediates could possibly represent up to 5 % of the overall mole fraction at high temperature flame but they are not detectable by Raman scattering.

An attractive approach which uses a combination of polarised and depolarised Rayleigh signals reported by [50, 51, 74, 101] which were performed to measure mixture fraction, temperature and molecular concentration is seen to possibly overcome most of the issues discussed earlier. Most of the scattered light from the molecules is polarised parallel to the polarisation of the laser. However, some of the radiation from molecules that are not isotropic will become depolarised. By contrast, molecules that are isotropic such as noble gases (He, Ar etc.), and those that have so-called “spherical-top” molecular configuration mostly found in fuels (CH$_4$ is one of them) will not produce any depolarised signal. This depolarised Rayleigh signal is usually neglected because the fraction is as small as ±1 % of the overall signal [63]. The depolarisation ratio of each species, $\rho_p$ is defined by the ratio of depolarised, $S_{dep}$ to the polarised intensity, $S_{pol}$ and in this work they were also taken from Fuest et al. [63] and it is read as in equation (3.6) where $\alpha$ is the mean molecular polarizability and $\gamma$ is the mean molecular anisotropy.

$$\rho_p = \frac{S_{dep}}{S_{pol}} = \frac{3\gamma^2}{45\alpha^2 + 4\gamma^2}$$ (3.6)
By using a suitable fuel and oxidiser combination, flame temperature has been shown to successfully be determined by exploiting these two signals. The procedure to infer the temperature from these two signals is now discussed.

As shown in equation (3.5), knowledge of the effective Rayleigh cross-section is required to accurately determine the temperature. As the Rayleigh measurement is itself not species specific, this could not be achieved experimentally (without another complementary measurement). An alternative approach to obtain species information is to perform numerical calculations of the combustion of the chosen fuel/oxidiser mixture and then calculate the effective Rayleigh cross-sections (for the polarised and depolarised components) using equation (3.1), as a function of a reaction progress variable (e.g. mixture fraction). The combined polarised and depolarised signals can then be used to identify the relevant reaction progress variable value, which allows the effective cross section to be recovered. Employing this more accurate effective cross section in equation (3.5) with the polarised Rayleigh signal delivers temperature. This method will be discussed in much greater detail in Chapter 5.

### 3.1.3 Laser Induced Fluorescence

The beauty of planar laser-induced fluorescence (PLIF) technique to extract species specific information in combustion has proven useful in combustion studies. Hydroxyl (OH) is one of the intermediate species present in combustion and usually regarded as a flame front indicator. The other intermediate species that has gained much attention from the combustion community is formaldehyde (CH\textsubscript{2}O) which has been a great indicator of cool-flame region that is generated in hydrocarbon oxidation process at low temperature region and further helping continuing the flame chemistry processes [102]. Both of these species are studied in this work, which the detail of specific measurement setup is explained in Chapter 4 and 5 as this section is only intended to provide a general description of the LIF technique.

Considering a two-level fluorescence model in Figure 3-1 the atom or molecule will be excited from ground level \(E_1\) to the upper level \(E_2\) of energy state, resulting from the absorption of photon energy stimulated by the laser source. The atom or molecule in this state is unstable, and may undergo different processes to return to the ground state by
releasing the extra energy through: (1) stimulated emission \((b_{21})\) due to the interaction of the incoming photon with the excited atomic electron which causes the atom return to the ground state, (2) photoionisation \((W_{2i})\) as a result of further absorption of photon, hence exciting it to a higher energy level, (3) predissociation \((P)\) through the interaction between atoms and molecules which dissociate prior to the emission of a photon, (4) quenching \((Q_{21})\) caused by the energy transfer between atoms or molecules via collision, and (5) fluorescence \((A_{21})\) due to the collision of molecule which causing it to return to vibrational level of the ground state, releasing energy in a form of fluorescence. To successfully image (depending on the measurement criteria either qualitative or quantitative) and determine the concentration of the intended species, several requirements need to be satisfied; first, every molecule has its own absorption spectrum, so the laser wavelength to excite that molecule is required. Secondly, as the fluorescence occurs at a different wavelength (usually longer) the emission spectrum of the molecule needs to be known so that an appropriate filter(s) could be used for the light collection. The rate of radiative decay of excited state, which directly influences the fluorescence signal, need also be known. Finally, the other process that competing with fluorescence such as predissociation and quenching also need to be taken into account which is really critical if one wants to get a quantitative measurement. For more detailed theory and physics information about LIF can be found in [103, 104].

![Figure 3-1: Energy level diagram for a two-level fluorescence model adapted from [5]](image-url)
Referring to [52] and based on the two-level energy diagram in Figure 3-1, rate equations for the population in the two energy levels; the excited state, $E_2$ and the ground state, $E_1$ can be expressed as

$$\frac{dE_1}{dt} = -E_1 b_{12} + E_2 (b_{21} + A_{21} + Q_{21})$$ (3.7)

$$\frac{dE_2}{dt} = E_1 b_{12} - E_2 (b_{21} + A_{21} + Q_{21} + P + W_{21})$$ (3.8)

With the assumptions that the steady state (ss) is achieved faster than the laser pulse (ss<< 10 ns), laser irradiance, $I_\nu$ is smaller than saturation limit, $I_\nu$ ($I_\nu \ll I_s$), predissociation $P$, photoionisation $W_{21}$, stimulated emission $b_{21}$ is negligible, the fluorescence signal, $F$ is proportional to $E_2 A_{21}$, and it can be expressed (details can be found in [26]) as

$$F = h\nu \frac{\Omega}{4\pi} l A_1^0 B_{12} I_\nu \frac{A_{21}}{A_{21} + Q_{21}}$$ (3.9)

where $h\nu$ is the photon energy, $B_{12}$ is the Einstein coefficient of absorption, $E_1^0$ is the Boltzmann fraction, $\Omega/4\pi$ is the portion of sphere of solid angle of the signal collection from the measurement area, $A$ and $l$ are the focal area and axial length of the fluorescence observation region respectively.

### 3.1.3.1 OH-PLIF

For the OH-PLIF experiment, a tunable dye laser (Rhodamine 6G solution in ethanol) pumped by an Nd:YAG laser is employed to excite the electronic state transition of $A^3\Sigma^+ - X^2\Pi$ (1,0). The absorption line (specific rotational line) however should be chosen after carefully considering the variation of the ground-state Boltzmann fraction over the expected temperature range. This is because the fluorescence intensity depends on the number of molecules available to be excited from a particular ground state. An absorption line which is very sensitive to temperature should usually be avoided although it might not be a problem if one performs a simultaneous OH and temperature measurement as the temperature
dependence on the Boltzmann population can be corrected, however keeping the least variation as possible would be ideal.

Figure 3-2 shows the Boltzmann fraction over the temperature range of 1000 °C – 2500 °C at 1 atm for several absorption lines that is used in several studies $Q_1(6)$ [69], $Q_1(7)$ [105], $Q_1(8)$ [106], and $Q_1(9)$ [107], calculated using LIFBASE. For a lower temperature measurement, the $Q_1(6)$ is favorable as it has a higher strength as shown Figure 3-3 and at the same time minimizing the effect of the thermal state population variation (~5 %). This is not the case in a high temperature combustion as the oxidation zone would be around 1500-2500 °C, and thus only two lines could be considered, i.e. $Q_1(8)$ and $Q_1(9)$ which giving the variation of 5 % and 8 % respectively, as the other lines contribute to more than 10 %. Considering laser linewidth of 0.06 cm$^{-1}$, the $Q_1(9)$ however overlaps with the $Q_2(8)$ and this should also be taken into account if this line is used. Quantification of OH concentration is possible as OH-collider quenching data is well documented [61] for major species involved in combustion. The quenching correction for OH thus becomes easy to implement if the major species and temperature are known.
3.1.3.2 CH$_2$O-PLIF

For the CH$_2$O measurement, rotational transition within a strong vibrational band, A$_2$A$_1$←X$_1$A$_1$ pQ can be excited using a laser wavelength ranging from 352 nm to 357 nm. This could be achieved using frequency tripled Nd:YAG laser at 355 nm [71, 108] or at 353 nm using excimer laser [109] or tuneable dye laser [110]. Some other excitations were also reported successfully at 370 nm hot-band excitation [111] and at 339nm of 2$_3^1$4$_0^1$ vibronic band [112]. The excitation of third harmonic Nd:YAG becomes favourable because of the availability of high pulse energy at this wavelength which compensate the overlapping at weak rotational transition of molecules [108]. This high pulse power also helps to saturate the PAH signal which was shown diminished at around tenth of mJ, but one has to ensure the CH$_2$O is still in the linearity region. The CH$_2$O has been reported to be able to maintain its linearity 100 times intensity higher than OH performed in the same conditions of flame and laser [113]. The CH$_2$O collision cross-section data is not so well established as for OH. Hence, a semi-empirical temperature dependent correction has been used in the past for semi-quantification. This correction considers the population distribution and quenching corrections proposed by Kyritsis et al. [114] and Paul and Najm [62] respectively. For the population distribution, the model takes into account the vibrational and rotational contribution using the Boltzmann fraction $f_0 = f_{\text{elec}} f_{\text{vib}} f_{\text{rot}}$. The electronic state however is
neglected. The quenching rate was said to follow a functional form somewhere between $Q_{12} \sim T^{-0.5}$ and $Q_{12} \sim T^{-1}$.

### 3.2 Experimental equipment

#### 3.2.1 Burner and flow configuration

All the experiments that will be explained in the next chapters were done using a simple non-premixed jet burner as shown in Figure 3-4 (a). This type of burner was chosen to enable the interaction behaviour between the oxidiser and fuel of oxy-fuel mixtures to be investigated. The burner is made from stainless steel, consisting of two co-flows, the inner (co-flow 1) with a diameter of 95.5 mm which surrounding the jet nozzle and the outer (co-flow 2) with a diameter of 211.56 mm. The gases that flowing through this burner has to flow through a series of perforated plates, with glass beads placed between them and then

![Figure 3-4](image)

*Figure 3-4: A schematic diagram of the non-premixed jet burner (a) and a photo of the burner secured to an optical table and placed on top of a laboratory jack (b).*
through a 100 mm height of honeycomb so that the gases are evenly distributed and uniformly flowing at the outlet. For the jet nozzle, a small stainless steel tube that has an inner diameter of 5 mm and length of 100d was used with the latter chosen to ensure the flow is fully developed. This nozzle is placed at the centre of the honeycomb and protrudes 40 mm from the burner exit.

Using this burner, two flame arrangements can be achieved: 1) Normal diffusion flame (NDF) which the fuel and the oxidiser is flowed through the jet nozzle and co-flow 1 respectively. Air, with the same velocity of co-flow 1 was flowed through the co-flow 2 to act as a shielding flow avoiding any entrainment to the measurement area of interest. 2) Inverted diffusion flame (IDF), which the fuel and oxidiser arrangement in 1) is swapped, meaning that the fuel is flowed through the co-flow 1 and the oxidiser is flowed through the jet nozzle respectively. The gas flowing through the co-flow 2 however is replaced with nitrogen to avoid a second flame generated a result of interaction between the fuel and air. This is important as the second flame would mislead the measured data (e.g. flame edge, mixed species generated by ‘oxy-fuel’ and ‘air-fuel’ combustion). The burner is secured to the optical table and is placed on top of laboratory jack as shown in Figure 3-4 (b) so that the burner can be easily moved up and down for the data collection at different heights.

Figure 3-4: Illustration of equipment and gas lines arrangement for experiment in discussed Chapter 4
The mass flow of all gases were set were set by a set of mass flow controllers, which was controlled by in-house Labview program. Figure 3-5 shows the schematic diagram for the flow arrangement that was used for experiment in Chapter 4, which the fuel and oxidiser mixtures were already mixed by the gas supplier. For the fuel and oxidiser mixtures in Chapter 5 and 6 however, the fuel and oxidiser mixtures were mixed in-house and so additional of two mass-flow controllers were employed to achieve this which two were used to mix the fuel and another two for the oxidiser.

The arrangement shown in Figure 3-5 also enables the flame swapping between the NDF and IDF to be done. This can be done by switching on the valves in the red dotted box to flow the fuel into the co-flow gas line while the oxidiser into the jet nozzle gas line. It should be noted that the helium and argon cylinders line was also connected to the jet nozzle and co-flow 1 line that was used for stray light and polarised light leakage imaging for the Rayleigh signal calculation that will be explained in Chapter 4 and 5. A solenoid valve is connected to the fuel line which is connected to a control box for safety reason, as such if the temperature drops below certain level or the emergency button is pressed, the solenoid valve is activated and the fuel line is stopped.

3.2.2 Lasers

Laser is an acronym for light amplifications by stimulated emission of radiation and has a radiation range from 1 nm to 1000 μm. This large spectrum can be divided into 5 different wavelength categories, namely deep ultraviolet (<200 nm), ultraviolet (200 to 400 nm), visible (400 nm – 700 nm), near infrared (700 nm - 10 μm) and far infrared (>10 μm). In combustion research, laser radiation in the ultraviolet (UV), visible (Vis) and near infrared spectra are always used. In this work, the laser wavelengths at 284, 355 and 532 nm are of interest, and the lasers used to produce this wavelength will be briefly explained.

3.2.2.1 Nd:YAG lasers

The Nd:YAG laser with its neodymium-doped yttrium aluminium garnet crystal (Y₃Al₅O₁₂) as a gain medium is widely used and usually pumped by flashlamps. In this type of laser, a four-level system, started with the excitation of doped neodymium ion (Nd³⁺) to the ⁴F₅/₂, and then followed by the relaxation of the ion to the ⁴F₃/₂ through non radiative processes, and
finally decaying to $^4F_{11/2}$ through lasing is achieved to produce a wavelength at 1064 nm. There are however other emission lines such as 1319 nm ($^4F_{3/2} \rightarrow ^4F_{13/2}$), 1123 nm ($^4F_{3/2} \rightarrow ^4F_{11/2}$), and 946 nm ($^4F_{3/2} \rightarrow ^4F_{13/2}$), but the 1064 nm has the best efficiency.

A high energy, pulsed Nd:YAG laser can be achieved through the use of Q-switching technique. In this technique, a large population inversion is created using the stored intra-cavity losses usually using a crystal known as Pockels cell, acting as a time-gated polariser, switches on or off the losses in the cavity.

In the Rayleigh scattering technique, or to use as a pump for dye laser, a laser wavelength at 532 nm is frequently used. The fundamental wavelength of Nd:YAG laser at 1064 nm can be frequency doubled using non-linear crystals such as beta-barium borate (BBO) or potassium dideuterium phosphate (KD*P) to achieve that. A 355 nm and 266 nm of laser wavelength can also be achieved from the fundamental frequency through the frequency triple and quadruple respectively.

At Imperial College, there is a unit of multi-YAG laser which houses four Nd:YAG lasers. This laser has high output energy which could achieve up to 1.8 J/ pulse (at 532 nm) by means of overlapping all the beams into one. To overlap beams that has same wavelength is hard to achieve. However, this system uses a trick by overlapping a different wavelength, one from the frequency doubled, and another one from the fundamental wavelength using a

![Figure 3-6: Illustration of Thales Multi-YAG laser system in 532 nm and 355 nm configuration used in Chapter 5](image-url)
combination of dichroic mirrors as shown in Figure 3-6. First, the fundamental wavelength of the laser 1 at 1064 nm (red line) was frequency doubled, generating 532 nm and a bit of 1064 nm. These two wavelengths were first separated using a dichroic mirror that transmits 1064 nm and reflect the 532 nm. Then, another dichroic mirror is used to combine the frequency doubled from Nd:YAG 1 onto the fundamental wavelength of Nd:YAG 2. As the phase matching of the doubling crystal only works at 1064 nm, the 532 nm can passes through while only the Nd:YAG 2 1064 nm is frequency doubled. The same process is repeated for all lasers if one wants to combine all the beams.

The wavelength of 266 nm in this laser can be achieved by placing the fourth harmonic generator (FHG) at the output of the overlapped 532 nm beams, or can also be done as in the 1064 nm to 532 nm conversions, by placing the FHG after the SHG and use dichroic mirrors that reflect 266 nm while transmit 532 nm and 1064 nm.

For the wavelength at 355 nm, this can only be achieved using a mixture of 1064 nm and 532 nm before this can be frequency triplet using the third harmonic generator (THG). This means that each of the line has to be frequency triplet at each line and combine them using dichroic mirrors as in the frequency double.

For the work that will be presented in the next chapters, the Thales multi-YAG laser were used in Rayleigh scattering experiments. The molecules were irradiated using overlapped beams from the first three cavities while the fourth cavity was used to generate UV light for LIF measurements (either pumping a dye laser or as the third harmonic).

### 3.2.2.2 Dye laser

The Nd:YAG laser can only have fixed laser wavelengths as explained earlier. Some molecules have a specific transition to be excited that cannot be achieved using the solid state lasers due to discrete energy levels of the gain medium. This is true especially in the application of LIF technique. The LIF technique also requires a narrow band laser system to avoid exciting other nearby transition lines. A dye laser can be used to overcome this problem, where organic dye molecules in solution are used as the gain medium (e.g. Rhodamine 6G dissolved in ethanol in case of OH-LIF). There are many selections of dye
depending on the application and the wavelength desired. This solution is continuously circulated to avoid dye degradation as well as to avoid triplet absorption.

A solid-state laser or flashlamp can be used to pump the dye molecule to achieve the population inversion. A frequency doubled Nd:YAG laser at 532 nm or frequency tripled at 355 nm is usually used as it is widely available. The repetition rate of the dye laser will be the same as the pump laser, and the energy used to pump the laser depends on the laser dye output desired, but mostly the dye laser can only generate 10 mJ/pulse and there should also be a maximum pump laser energy allowed which usually below than 600 mJ.

The wavelength of the dye fluorescence is determined by the angle of tuning mirror to the grating. For example, in OH-LIF experiment, the OH atom is usually excited at around 283 nm and so the emission from the dye laser has to be tuned at around 566 nm before frequency doubled. For the work that will be presented in the next chapters, a Sirah PrecisionScan dye laser was used to excite the OH atom.

### 3.2.3 Light detectors

Light detectors collect all the information by means of collecting electrical charge converted from photons based on photoelectric effect. These light detectors might have the same physical principle however the sensor and architecture differentiate the use of one type of camera to another and so depending on the measurement of interest and the requirements, the camera must be carefully chosen.

There are two main aspects that should be considered. First is the amount of light that will be collected. For example in OH-LIF and CH$_2$O-LIF imaging, the amount of light produced is usually small as it depends on the excited species concentration in the flame. This motivates the use of an intensified charge coupled device (ICCD) for the LIF technique while the CCD is more than enough for the Rayleigh measurement. There are also different types of photocathode and phosphors that are used in the ICCD, which the sensitivity of different phosphors/photocathodes depends on the wavelength of the light collected. Second is the minimum exposure time of the camera. In CCD camera, the exposure time depends on how fast the readout can be achieved. The minimum exposure duration of a CCD camera is usually around 1μs. For an ICCD, the intensifier module can gate the exposure to 100 ns or
less, which can be useful for background suppression and/or elimination of elastic light scattering. Exposure/gate time is important in turbulent imaging as the flow has to be frozen (i.e. stationary during acquisition) and so the minimum possible exposure time is definitely required. Proper synchronisation tests between the CCD, ICCD and laser pulse have to be performed to ensure the timings between the equipment are correct.

The CCD and ICCD camera that are used in this work will be explained next.

### 3.2.3.1 CCD cameras

The conversion of the photon into electric charge is done using a CCD sensor, which every camera has a number of elements (known as pixels) with a size of 6.45 μm x 6.45 μm each. The arrangement of the pixels in an array enables the read out of electron to be done by transferring the electron, one at a time vertically to the bottom edge of the sensor which is known as analog shift register. This electron will be then transferred through the charge-to-voltage converter, where the collected amount of charge from each pixel is proportional to the voltage.

The sensors can be arranged in different ways namely full frame transfer, frame transfer, interline transfer and also full frame interline transfer. The full frame transfer and frame transfer are highly affected by charge smearing. This happens because when the charge signal is being readout, the incoming photons could fall onto the sensitive sensor array. This smearing effect can be avoided by employing the interline transfer CCD. In interline transfer sensor, every column of pixels has its own vertical transfer register next to every column of the active pixels and are shielded to avoid exposure to light. The charge that falls onto each sensor can be rapidly transfer to the vertical transfer register, usually in order of less than 1 μs before being readout, and at the same time the exposed active pixels are reset and a new data collection can be done. In this work, the interline transfer sensor cameras (LaVision Imager Intense) were used. The (polarised) Rayleigh signal was collected using this camera alone, while for the depolarised Rayleigh, OH-PLIF, and CH₂O-PLIF signals, the cameras were coupled with intensified relay optics (IRO) devices.
3.2.3.2 Intensified CCD cameras

ICCD consists of a CCD camera and an intensifier, where the latter is mounted in front of the camera. ICCD is very useful to be used in the application where the incident light is really low, where the intensifier is used to amplify the signal before being transferred to the phosphor screen and finally onto the CCD sensor using coupling lens. The image intensifier consists of three functional units namely photocathode, multi-channel plate (MCP) and phosphor screen. Photons from the measurement area will first arrive at photocathode where it is placed behind the entrance window. Entrance windows made from quartz are usually used, but if the ICCD is used to collect a transmission in deep UV, MgF₂ window can be employed. The quantum efficiency of the conversion of the photons to photo electron depends on the photocathode material as well as the incoming light wavelength. Figure 3-7 shows the quantum efficiency versus wavelength of different material of the photocathode. For example, if one wants to use the ICCD in the visible spectrum, GaAsP photocathode has a very high efficiency compared to GaAs, however it has a smaller wavelength spectrum. The S20 photocathode is known one of the most popular photocathode as it covers a wide spectrum from UV to near infrared.

The electrons from photocathode are then passed through the MCP where the electrons are then multiplied. The MCP can be either single stage, double stage or even triple stage. For the single stage, 1000 secondary electrons are produced per incoming photon, while in double stage, $10^6$ secondary electrons can be achieved. However, maximum multiplication that can be gained from triple stage is limited only to $10^8$ per incoming photon to avoid saturation.

The phosphor screen, being the last functional unit will release photons when the electrons accelerated from the MCP hit the screen. Different types of phosphor indeed play a major role in determining the conversion factor of the phosphor screen. Widely used phosphor screens are P43 and P46. The P46 has a higher quantum efficiency compared to the P43, however the phosphor decay time is longer compared to the latter. A longer decay time phosphor should be avoided in a high speed application as it would produce ghost image as well as cross talk to the next image. Table 3-1 are the IROs list that were employed in the
experiments performed in this work. All the IROs use a lens-coupled MCP to intensify the image.

![Quantum efficiency vs wavelength of various photocathode](image)

*Figure 3-7: Quantum efficiency vs wavelength of various photocathode adapted from [3]. SBA is super bialkali and UBA is ultra bialkali*

<table>
<thead>
<tr>
<th>Collected signal</th>
<th>Detection Wavelength (nm)</th>
<th>Phosphor screen</th>
<th>QE %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rayleigh (depolarised)</td>
<td>532</td>
<td>P43, GaAsP</td>
<td>60</td>
</tr>
<tr>
<td>OH-PLIF</td>
<td>308</td>
<td>P46, S20</td>
<td>14</td>
</tr>
<tr>
<td>CH$_2$O-PLIF</td>
<td>400-500</td>
<td>P43, S20</td>
<td>15</td>
</tr>
</tbody>
</table>

*Table 3-1: Specifications of IROs used in the experiment*
4 Simultaneous Rayleigh and OH-PLIF signals

The result in this chapter is published in Flow, Turbulence and Combustion under a title of ‘A Combined Experimental and Numerical Study of Laminar and Turbulent Non-piloted Oxy-fuel Jet Flames Using a Direct Comparison of the Rayleigh Signal’ (pages: 70 – 80) and in Proceedings of the Combustion Institute under a title of ‘Comparative Flame Structure Investigation of Normal and Inverse Turbulent Non-premixed Oxy-fuel Flames Using Experimentally Recorded and Numerically Predicted Rayleigh and OH-PLIF Signals’ (pages: 81 - 83). In this work, I performed all the experiments, analysed and proposed the fuel/ oxidiser mixtures and performed data post-processing. The numerical simulations were performed by Franziska Hunger from Freiberg University, Germany. For the journal papers, the experimental part was written by me, and the numerical part by Ms Hunger. The other sections, figures, and analysis are a joint effort between myself and Ms Hunger with advice from Dr Ben Williams, Professor Frank Beyrau and Professor Christian Hasse. For the sake of clarity, please note that the numerical results in this chapter are presented in order to complete the story about the numerical/experimental comparison and I do not claim sole intellectual ownership.

4.1 Introduction

In this chapter, direct comparison of experimental and numerical Rayleigh and OH-PLIF signals of laminar and turbulent non-premixed oxy-fuel flames are investigated. The experimental result in this chapter was used to validate the oxy-fuel flame numerical modelling. Oxygen-enhanced oxy-fuel flames exhibit different behaviours compared to more typical CH₄-air flames, and hence better understanding of the structure of this kind of flame is required. Furthermore, altered chemical paths due to the added flue gas (CO₂), high heat losses, high flame temperature and high possibility of soot formation require different treatment in terms of diagnostic techniques as compared to CH₄-air flames, and this will also be addressed here.

Commonly, experimental signals need to be post-processed to infer the physical quantities (e.g. temperature, mass fraction) before numerical validation can be done. The assumptions made in the post-processing ‘stage’ however often leads to inaccuracies. Considering the
Rayleigh scattering technique for temperature measurement for example, this technique requires a constant effective Rayleigh cross-section throughout the flame to directly convert the Rayleigh signal into temperature. However only certain fuel/oxidiser combinations have been shown to able to keep the inaccuracy in an acceptable experimental level as investigated by Bergmann et al. and Dibble et al. [45, 46]. This inevitable inaccuracy becomes pronounced in more complex fuel/oxidiser mixture.

A simple experimental arrangement together with high signal to noise ratio (SNR) in Rayleigh measurement in combustion research have led some investigation to create the possibility of employing this technique for the study in broader fuel/oxidiser mixture. In an investigation performed by Stepowski and Cabot [115], they demonstrated the possibility of generating temperature and mixture fraction data by employing algorithm to process the Rayleigh signal in H2-air flame. This algorithm requires the use of a strained flame library. However, many assumptions are required, which again lead to a significant uncertainty when applying in complex fuel/oxidiser especially in oxy-fuel.

A complex fuel/oxidiser mixture has been presented by Fuest et al. [63] where combined Raman/Rayleigh scattering techniques were employed to measure species compositions and the mole fractions were then used to measure the effective Rayleigh cross section to infer the temperature quantity in DME flame. By considering only main species and neglecting lower concentration species (OH, H, and O), the contribution to the Rayleigh signal was assumed as a collective of eleven species (CO2, O2, CO, N2, H2O, H2, DME, CH4, C2H4, C2H6, and CH3). The Raman signal collected from the experiment however composed only six non-hydrocarbon species and a model was used to estimates the remaining five species (hydrocarbon species) to determine the effective Rayleigh cross section which was then use determine the temperature. The contribution of the species that were neglected in their experiment composes more than 6% in oxy-fuel flame rendering this approach ineffective for the current work.

Connelly et al [116] presented an idea to reduce the uncertainty by comparing experimental and numerical data through directly measured signals rather than physical quantities. The beauty of this approach lies on fewer, simpler measurements as well as a higher signal-to-noise ratio of the data. NO-LIF using both traditional (physical quantity) and signal
comparisons method were investigated in sooting and non-sooting diffusion flames, and concluded that similar quantitative information was obtained using both methods, but reducing uncertainty of course, for the direct comparison of the signals. This approach later was used by Coriton et al. [117] for the OH- and CH$_2$O-LIF signals in turbulent DME flame. A comparison of polarisation/depolarisation Rayleigh signals to numerical result was then investigated by Shießl et al. [118], with the emphasis of determining the species and temperature using Newton iteration in turbulent, non-premixed, Sandia flames signals. It has been shown that the planar map from the measured Rayleigh signal can be used to map several species and also temperature using the computed polarised and depolarised signals.

Realising the ability of greatly reducing uncertainty by applying the experimental and numerical direct signal comparison approach, the idea is extended to investigate laminar and turbulent oxy-fuel flames structure in non-premixed jet burner by employing the Rayleigh scattering and OH-PLIF techniques. Rayleigh scattering was preferable as the signal generated represents all the existing species, as well as making a quantitative validation across the field of view feasible, which has a great advantage over other techniques such as OH-PLIF which only generated in flame front as well as limited to qualitative measurement, or Raman scattering which only limited for 1D measurement, and only available for major species.

The first comparison of this investigation was done in laminar flame due to the availability of fully resolved numerical species and temperature profiles. Through this comparison, a detailed study was performed by analysing the different modelling approaches, assumptions in diffusion as well as radiation models. Then, the second part involves the statistical comparison of the turbulent flame by comparing the time average, variance or RMS and probability density function (PDF) of the evaluated Rayleigh signals. Finally, a simultaneous measurement of Rayleigh and OH-PLIF signals was performed to study in detail the structure of normal and inverted oxy-fuel flames and the results are compared to the numerical simulation. The Rayleigh ratio signal was used to quantitatively validate the overall structure of the field of view, while the OH-PLIF used to qualitatively indicate the flame front structure. Simultaneous measurement enables further analysis to understand the correlation between these two signals. Furthermore, as two signals were used to validate the numerical simulation results, this will increase the confidence in the numerical model.
reliability, as well as proving the feasibility of applying direct signals comparison technique in combustion research.

4.2 Methodology

4.2.1 Boundary conditions

The boundary conditions for both experiment and numerical setups in a cold flow were first investigated to ensure a match and avoid the propagation of inaccuracies into the calculated domain. This measurement and calculation were performed as a function of radial locations at 1 mm, 3 mm, 10 mm and 20 mm above the nozzle tip. The hot wire probe was traversed in steps of 0.5 mm at +/-10 mm from the nozzle centre and 1mm step separation beyond that. The measurements were taken by flowing air through the jet nozzle at Re=18000 (jet mass flow rate = 1.0264 x 10^3 kg/s) and co-flow velocity of 1 m/s. The evaluation was first made to confirm the accuracy of the mass flow controllers by measuring the bulk velocity

![Figure 4-1: Comparison of mean and RMS axial velocity from the hot wire measurement and numerical calculation at Re=18000 at 1, 3, 10 and 20 mm above the nozzle tip](image)
(29.7 m/s) by integrating the HWA data, and the result produced stems to the calculated velocity (29.3 m/s) from the mass flow controllers.

The numerical jet velocity inflow boundary was based on the pipe flow simulation, aiming to generate turbulent time-dependent inflow profiles simulated with an in-house solver based on the OpenFOAM® 2.1. The profiles calculated in the jet boundary were then transported into computational domain. The co-flow velocity however was set to a block profile. A rectilinear computational domain with a grid resolution of 64 x 64 x 300 cells with grid size of 30 x 30 x 150 mm and 60 x 60 x 150 mm at the jet inlet and outlet respectively were used. The statistics results presented in this section are the averaged of simulated value for three flow through times (FFT) for a period of 12 FFT by time-integration.

The mean and RMS measurement values at the different radial locations were then compared between the measured and numerical calculation. Figure 4-1 shows the mean and RMS axial velocity from the hot wire measurement and the numerical calculation. The mean velocity at all radial locations agree well for both the numerical and experimental results. However, for the RMS velocity, only the RMS at 3 mm agrees well to each other while the numerical calculation underestimates at radial location of 1 mm, and overestimate at 10 mm and 20 mm above the nozzle. The most important aspect is the structure of the flow fields, and they are captured well thus the boundary conditions were considered to match.

4.2.2 Fuel and oxidiser selection

A stable flame that can be operated at high Reynolds numbers is required to enable a comparison of normal and inverted turbulent flames in the non-premixed burner. At least 30 % of O₂ (for O₂ and CO₂ oxidiser mixture) is required to achieve air-like combustion stability as reported by Diranto and Hals [94]. This stability can further be improved with an addition of H₂ in the fuel stream which helps increasing the global reaction rate of the flame, compared to CH₄ alone which has a relatively low chemical reaction rate. This low chemical reaction rate coupled with high turbulent mixing [57] could possibly induce localised flame extinction, which lead to lift-off or blow-off. This becomes more critical when dealing with the inverted flame as it has a narrower stability limit compared to the normal flame configuration.
Coupling the aforementioned requirement to operate the flame at a high Reynolds number with the main objective to study an oxy-fuel flame structure at an elevated temperature (>2400 K) necessitates tailoring a suitable fuel and oxidiser combination. A numerical simulation of opposed laminar flame was performed using CHEMKIN-PRO [119] at a strain rate of 100 s⁻¹ to understand the minimum O₂ volume fraction needed to achieve this temperature of interest. From the calculation, 68 % of O₂ in oxidiser stream is required. This huge amount of O₂ is required mainly due to the cooling effect generated by CO₂ as it has a higher heat capacity compared to the N₂, as well as radiative heat loss which accounted four times higher than equivalent N₂ flame as reported by Samaniego [120].

The other important aspect that was considered is the soot generated by the mixture, as it has to be kept at a minimum level as possible to avoid any contamination of the Rayleigh signal. The soot generation is at its peak between 30 %–40 % O₂, and further decrease with increasing of O₂ in oxidiser stream due to promotion of soot oxidation as reported by Wang et al. [79] in their investigation in propane flames. As explained earlier in the previous

![Graphs showing normalised Rayleigh cross section, normalised Rayleigh signal, mole fraction at 1000 s⁻¹, and temperature from the laminar opposed flame calculation at different strain rates for (fuel: 17.5 % CH₄, 40 % CO₂, 42.5 % H₂, oxidiser: 68 % O₂, 32 % CO₂).]
paragraph, this will not be a problem considering the O\textsubscript{2} content in the oxidiser stream accounts for a minimum 68 \% to achieve the proposed temperature. The existence of the CO\textsubscript{2} in the fuel/oxidiser mixture at high temperature has also been reported by Mazas et al. [121] and Du et al. [122] decreasing the H radicals which helps in reducing soot formation through the reaction of CO\textsubscript{2} + H \leftrightarrow CO + OH.

A significant production of radicals (OH, O and H) in high temperature oxy-fuel flames gives a huge impact in maintaining the Rayleigh cross section throughout the combustion process (from reactants to products). The Rayleigh cross section variation also becomes worse due to the dissociation of CO\textsubscript{2} into CO in this present oxy-fuel case which generally happens at a high temperature flame. An appreciable variation after an attempt in maintaining the cross section cannot be neglected, and thus the direct conversion from the Rayleigh signal into the temperature space as performed by [46]and [45] whose variation is only 3 \% and 5 \% respectively is impossible.

After considering all the requirements as explained above, an optimised fuel/oxidiser combination was found to be (fuel: 17.5 \% CH\textsubscript{4}, 40 \% CO\textsubscript{2}, 42.5 \% H\textsubscript{2}, oxidiser: 68 \% O\textsubscript{2}, 32 \% CO\textsubscript{2}). Figure 4-2 shows the laminar flame calculation of opposed jet flame using CHEMKIN-PRO employing GRI-MECH 3.0 for the investigated fuel and oxidiser combination.

The estimated normalised Rayleigh cross-section (flame cross-section divided by oxidiser cross-section), Rayleigh ratio (oxidiser signal divided by flame signal. This will be explained in greater detail in section 4.3. See equation 4.6) and temperature of a range of strain rate (100 s\textsuperscript{-1} – 1000 s\textsuperscript{-1}) that is expected in this flame were calculated and presented. The mole fraction at a strain rate of 1000 s\textsuperscript{-1} is also presented in the figure above. The stoichiometric mixture fraction of this flame was found to be 0.418. The normalised Rayleigh cross-section (top left), \( \sigma_{\text{Ray,eff}} \) of the flame divided by the \( \sigma_{\text{Ray,eff}} \) of the oxidiser was calculated using equation (3.1) with species-specific cross section \( \sigma_{\text{Ray,i}} \) values from [63] weighted by the respective mole fractions \( X_i \) taking into account 14 species (H\textsubscript{2}, H, O, O\textsubscript{2}, OH, H\textsubscript{2}O, CH\textsubscript{3}, CH\textsubscript{4}, CO, CO\textsubscript{2}, CH\textsubscript{2}O, C\textsubscript{2}H\textsubscript{4}, C\textsubscript{2}H\textsubscript{6} and C\textsubscript{3}H\textsubscript{8}) that are involved in the reaction. The calculated variation of the Rayleigh cross-section is between 5-15 \%. The peak fluctuation is seen to happen due to a high generation of the CO\textsubscript{2} in the lean side as shown in the mole fraction subplot (bottom left). However, this cross-section is decreased in the rich side due to the
dissociation of CO\textsubscript{2} into CO resulting from the high temperature. The peak of H\textsubscript{2}O is also seen in this region and as the combination of the Rayleigh cross section of the H\textsubscript{2}O and CO is smaller than the CO\textsubscript{2}, this produces a minimum value of the effective cross section at the peak of H\textsubscript{2}O and CO. The top right subplot shows the Rayleigh ratio reduces as the strain rate increases. The same behaviour was also noticed in the temperature subplot (bottom right), where the temperature decreases as the strain rate increases. Note that the Rayleigh ratio is proportional to the temperature.

Figure 4-3: Illustration of simultaneous Rayleigh and OH-LIF measurement setup
4.2.3 Experimental setup

The measurement of Rayleigh scattering and simultaneous Rayleigh and OH-PLIF used the setup as illustrated in Figure 4-3. For the Rayleigh measurement alone, the dye laser and ICCD were not used. A combined energy of spatially overlapped beams generating of about 1.8 J/shot produced by four frequency-doubled Nd:YAG laser heads (THALES Multi-channel system) was used to induce the Rayleigh scattering. Care was taken to limit the peak intensity through the harmonic crystals by operating the laser system in a burst mode, with four laser pulses separated by 30 ns, repeated at 10 Hz. A ½ wave plate was used to rotate the horizontal polarised beam to vertical orientation, and a combination of lenses were used to create a laser sheet with a thickness of 120 μm, and a height of 25 mm. An iris was then used to block all the stray light produced on the top and bottom of the laser sheet. The stray light was further reduced using beam stops and matte black screens. An un-intensified interline transfer CCD camera (LaVision Imager Intense) with 1280 x 1024 pixels was used to collect the Rayleigh scattering light. After 2 x 2 hardware binning, an imaging region of 33 mm x 25 mm was created with a geometric resolution of about 50 μm/pixel. This CCD camera was fitted with a camera lens (f/1.2, f=50mm) and a 532 nm interference filter (ΔλFWHM = 3nm). Minimum exposure time of the camera at (1 μs) was set to collect the light.

For the combined measurement, the same system for the Rayleigh scattering explained earlier was used, except only the first three of 532 nm laser beams from the laser system were spatially overlapped producing about 1.5 J/shot, while the fourth cavity was used to pump the Sirah PrecisionScan dye laser after a delay of 430 ns to avoid cross talk between the diagnostics. Despite the time delay, this joint measurement is still instantaneous because the elapsed time was orders of magnitude shorter than the flow timescale. Rhodamine 6G solution dissolved in ethanol was used to produce a light at 566 nm which was then frequency-doubled in temperature controlled BBO crystal. This was finally separated from the fundamental frequency by a four-prism separator and the beam was expanded and focused using a combination of spherical and cylindrical lenses. The Q1(8) line of the A2Σ⁺-X2Π (1,0) transition at around 283 nm was excited using 5 mJ/pulse which is well below the saturation regime. This excitation was chosen to minimise the ground-state Boltzmann fraction population distribution variation over the expected temperature range,
which a variation of below than 7 % was predicted using LIFBASE [123]. For the OH-PLIF, ICCD camera (LaVision imager intense plus IRO) with a 100 mm Halle AR-coated UV lens (f/2.0) was used to acquire the fluorescence signal from the $A^2 \Sigma^- - X^2 \Pi$ (1,1) and (0,0) transitions peaking at around 308 nm. Colour glass filters (1mm thick Schott UG5 and WG305) were used to block elastic scattering signal, and the intensifier was gated at 200 ns to reduce background luminosity. Before performing the simultaneous measurement, these two cameras were calibrated by imaging a transparent dots target, which is used in post-processing to realign and correction for distortion, with sub-pixel accuracy. Further correction of the energy fluctuation, dark noise, stray light as well as spatial uniformity of the images will be explained in Section 4.2.4.

### 4.2.4 Post processing

<table>
<thead>
<tr>
<th>Laser diagnostics technique</th>
<th>Rayleigh scattering measurement</th>
<th>Joint Rayleigh and OH-PLIF measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flow conditions</td>
<td>Turbulent</td>
<td>Laminar</td>
</tr>
<tr>
<td>Jet condition</td>
<td>Re=18000 (fuel)</td>
<td>Re=850 (fuel)</td>
</tr>
<tr>
<td>Co-flow velocity</td>
<td>1 m/s (oxidiser)</td>
<td>1 m/s (oxidiser)</td>
</tr>
<tr>
<td>Measurement locations (from nozzle tip)</td>
<td>23-48mm, 48-73mm, 98-123mm</td>
<td>23-48mm, 60-85mm</td>
</tr>
</tbody>
</table>

Table 4-1: Experimental conditions for Rayleigh and simultaneous Rayleigh and OH-PLIF measurement

As explained earlier in the previous subchapter, all the acquired images were corrected for the distortion, and for the joint Rayleigh and OH-PLIF measurement, image mapping algorithm was performed for every pixel in every image using algorithm available in Davis software. The mismatch as determined from the transparent dots target was in order of subpixel. Table 4-1 shows the experimental cases that were performed in the Rayleigh and joint Rayleigh/OH-PLIF measurement. Nine different experimental conditions: Three measurement locations for turbulent Rayleigh measurement and two measurement locations for each of laminar Rayleigh, NDF and IDF joint Rayleigh/OH-PLIF measurement...
were collected. Every condition consists of three (Rayleigh measurement) or four (joint Rayleigh/OH-PLIF measurement) data sets containing a minimum of 500-1000 images each. The three data sets for the Rayleigh measurement were 1) flame, 2) helium flow and 3) oxidiser flow images, while the joint Rayleigh/OH-PLIF images sets were: 1) flame, 2) helium, 3) nitrogen and 4) acetone. Finally, the camera dark current images were also collected by means of keeping the caps on while taking the images.

The data post-processing was started with the laser energy fluctuation correction for the OH-LIF. The fluctuation of the Rayleigh laser was measured below than 3 %, and the fluctuation was neglected. Then, based on the Rayleigh images, any image contaminated with particles was discarded.

For the OH-PLIF signal, as only qualitative measurement was of interest, the OH-PLIF signal was only corrected for the beam shape (and not the shot-to-shot energy fluctuation) using a mean image of laser-induced fluorescence collected from acetone vapour in a quartz-cell.

For the Rayleigh measurement, care was taken to ensure stray light was kept at a minimum level as possible. The laser beam was surrounded by a black tube, and some diffuse light at the top and bottom of the beam sheet was reduced by using an iris. A matte black wall which acts to absorb most of the secondary scatter of Rayleigh photons was placed around the measurement area to reduce the chance of stray light getting into the camera lens. However, a very small amount of stray light was still collected, and has to be subtracted from the recorded images. To account for this, the helium and oxidiser images were used to determine the amount of the stray light. Images forming the average for the oxidiser, helium and dark current were performed. The raw pixel signal, $S_{raw}$ read as [124]:

$$S_{raw}^{He} = S_{R}^{He} + S_{stray} + S_{dark}$$

$$S_{raw}^{Ox} = S_{R}^{Ox} + S_{stray} + S_{dark}$$

Where the $S_{R}$ is the molecule(s) Rayleigh signal, $S_{stray}$ is they stray light collected by the camera from the surrounding, and $S_{dark}$ is the dark current of the camera. With Rayleigh cross section $\sigma$ for both of these molecules are known, and Rayleigh scattering signal section is proportional to the scattering cross section $\sigma$, it follows
\[ S_R^{He} = \frac{\sigma^{He}}{\sigma^{Ox}} S_R^{Ox} \] (4.3)

The stray light contribution to the signal can thus be calculated using

\[ S_{stray} = \left[ (S_{raw}^{He} - S_{dark}) - \frac{\sigma^{He}}{\sigma^{Ox}} (S_{raw}^{Ox} - S_{dark}) \right] \left[ 1 - \frac{\sigma^{He}}{\sigma^{Ox}} \right]^{-1} \] (4.4)

In some cases, the helium signal can be directly used as the stray light contribution (without the need to get the stray light value from equations above) due to its small Rayleigh cross section, and hence almost negligible signal. However, in this experiment, as high laser energy is used to excite the molecules, the helium signal value is appreciable, and this helium scattered light cannot be ignored.

The final stage of the post processing was the normalisation of the flame image to the oxidiser average image to determine the Rayleigh ratio \( R_R \) using equation (4.5) for every flame image so that the comparison between the experiment and the numerical calculation can be done in a shot-to-shot basis. This at the same time cancels any non-uniformity in the laser sheet.

\[ R_R = \frac{S_R^{avg\ Ox}}{S_R^{flame}} = \frac{S_{raw} - S_{stray} - S_{dark}}{S_{raw} - S_{stray} - S_{dark}} \] (4.5)

It should be noted that the Rayleigh ratio, \( R_R \) is proportional to the temperature, which is opposed to the Rayleigh signal, \( S_R^{flame} \). For the Rayleigh signal, the signal is low at higher temperature due to reduced density (see equation 3.5). However, the Rayleigh ratio is proportional to the temperature (see Figure 4-2).

**4.3 Numerical Approach**

This subsection is not intended to explain the numerical approach in detail. Readers who are interested in a detail of explanation may refer to [125]. Only brief explanation of simulation procedure, modelling approach and numerical calculation for Rayleigh ratio are considered here in order to provide relevant background for the experimental/numerical comparison.
The laminar Rayleigh ratio signals from the experiment were first compared to four different kinetic mechanisms namely GRI-MECH 3.0 [126], reduced GRI-MECH 3.0 [127], and mechanisms by Rasmussen et al. [128] and Wang et al. [129]. The differences in these mechanisms are only the variations of the number of species and reactions considered. With a little influence to the fuel/oxidiser mixture considered in this investigation, GRI3.0 was employed.

Using the GRI-MECH 3.0 kinetics mechanism, the second study performing a comparison between several sensitivity radiation models to the experiment considering optically thin model, OTM (considers only radiative heat loss) and P1 (considers radiative and reabsorption heat loss) in laminar flame was done. The outcome of this study will be explained in section 4.4.1. All the simulations in laminar flames were performed using Freiberg University’s in-house solver based on OpenFOAM 2.1.

With the same in-house solver, the turbulent flame was simulated using flamelet/progress variable approach [130] within the Large Eddy Simulation framework. An initial test to study the influence of the differential diffusion effects in the turbulent flame at Re=18,000 near field where it is expected to be dominant were investigated. This is done by applying unity Lewis-number so that comparison with constant, but non-unity Lewis numbers can be performed. The result however showed by considering this fuel/oxidiser mixture, at the near field regions, unity Lewis number approach gives close approximation to the experimental result and the differential diffusion effects are less dominant.

The result of the numerical simulation is compared to the experiment in terms of Rayleigh ratio. From the simulation of this fuel/oxidiser mixture, all the local and instantaneous species coupled with temperature were used to calculate the numerical Rayleigh ratio, using equation (4.5). However the stray light and dark current do not exist in numerical space, so those variables were discarded and the Rayleigh ratio can simply be calculated as

\[ R_R = \frac{T}{T_{Ox} \sum_{j=1}^{n_{Flame}} X_j \sigma_{Rj}} \]  

(4.6)

Here, \( T \) is the local and instantaneous temperature, \( T_{Ox} \) is the oxidiser temperature and \( X \) is the mole fraction of the considered species. All the considered 14 species here make up
almost 99.5% of overall species involves in the reaction. The changes in cross section due to temperature dependencies was shown to be up to 2 % by [131], however only 1 % in DME flame investigated by [63]. An estimation of this effect was performed using the relationship suggested by Fuest et al., and it resulted of not more than 2 % of the cross section inaccuracy and no further correction was made.

For the OH-PLIF signals, the Q₁(8) line of the A² Σ⁺→X² Π \(1,0\) band in the linear LIF regime considering the excited-state decay rate dominated by collisional quenching was calculated using

\[ S_{OH-LIF} \propto N_{OH} f_B \frac{A}{A + Q} \]  \hspace{1cm} (4.7)

Where \( N_{OH} \) is the OH number density, Boltzmann fraction represented by \( f_B \), \( A \) is the spontaneous emission rate and \( Q \) is the total collisional quenching rate of the excited state. The total quenching rate that considers all collisional partners, where the quenching species of \( OH, CO_2, CO, CH_4, H_2, H_2O, O_2 \) and \( N_2 \) were calculated and correlation and quenching cross section for OH according to Tamura et al. [61] were used.

### 4.4 Results and discussion

This subchapter is divided into three sections. The first section is the comparison of laminar flame detailing the sensitivity study of radiation models and also the influence of Soret effect in this type of flame. This was done to fully utilise the fully resolved species and temperature data of laminar simulated flame. Contrary to the laminar flame, in turbulent flame, a direct side-by-side comparison is impossible to be done. This however can be achieved by comparing the statistics of both simulated and recorded data. The mean and variance or RMS data will be first presented, followed by the analysis on probability density function (PDF) in the second section. In the third section, a joint measurement of Rayleigh and OH-PLIF signals in normal and inverted non-premixed flames will be discussed.
4.4.1 Comparison of experimental and numerical Rayleigh signals in laminar flame

Figure 4-4 (a-d) shows a side-by-side comparison of the experimental and numerical results at two different locations in the laminar flame. The simulation considers different radiation models: OTM (considers only radiative heat loss), P1 (considers radiative and reabsorption heat loss) approaches as well as adiabatic. The radiation effect is not clearly giving much difference in term of Rayleigh ratio distribution between the P1 and OTM approaches at the upstream location. The adiabatic simulation however estimated a thicker flame, indicated by the high Rayleigh ratio (red region in (d)) compared to the experiment and the other two radiation models. In the downstream location, the overestimation from the adiabatic simulation is clearly seen, while the OTM radiation model behaved in opposite way by underestimate the Rayleigh signal. Structurally, P1 approach gives a very close estimation to the experimental result for both measurement locations. The quantitative comparison was then done by comparing the absolute Rayleigh ratio value between the experimental and numerical. For the upstream and downstream locations, lines at radial positions of 35 mm and 75 mm respectively are plotted as shown in Figure 4-4 (e-f) for this purpose. The experimental result was plotted as error bars indicating the fluctuation of the Rayleigh ratio data for the individual pixel. At the 35mm location, the P1 and OTM approaches sit in the experimental uncertainty from the nozzle centre until about 7.5 mm where the OTM underestimate the Rayleigh value beyond that while P1 approach continues giving a very close approximation to the experimental value. This is due to the consideration of both radiative and reabsorption heat loss in P1 approximation, as in oxy-fuel flame, high concentration of CO₂ and H₂O exist and both molecules absorb and emit radiation. Adiabatic simulation however gives overestimation value at the high temperature region (i.e. about 2.5 mm to 9.0 mm), but agrees well with the experimental data at low temperature regions (near the nozzle and oxidiser co-flow). At 75 mm, the Rayleigh ratio difference is more pronounced compared to the 35 mm. The adiabatic simulation clearly overestimates the Rayleigh ratio value while it is underestimated by the OTM approach. The P1 approach however yields a good agreement to the experimental data at high temperature region (from the nozzle centre to about 9 mm), while underestimates at the lean region. These overestimation and underestimation of the Rayleigh ratio values from the adiabatic and
OTM approaches are mainly affected by the difference of temperature estimation by these models, and less influenced by the species profile changes (please refer [125] Fig. 6).

The importance of considering the response of molecule towards the force of temperature gradient (Soret effect) is worth to study due to the combination of light (e.g. H\textsubscript{2}) and heavy (e.g. CO\textsubscript{2}) molecules in the fuel mixture used in this work. For this analysis, P1 radiation approximation approach was employed to study the Soret effect as it was shown to be closest to the experimental data as revealed in the radiation analysis earlier. Figure 4-5 (a-c) shows the side-by-side comparison of the experimental data, laminar flame simulation using...
P1 radiation considering Soret effect and without Soret effect respectively. The difference in the flame structure for both models in comparison to the experimental image at the downstream and upstream region is not clearly seen. To analyse this in detail, as in the radiation study earlier, a line at 35 mm and 75 mm are extracted from the images and plotted as shown in Figure 4-5 (d) and (e) respectively. At 35 mm location, both of the simulations agree well with the experimental data, while at 75 mm at the jet nozzle region the produced ratio are still in the experimental uncertainty, but deviate beyond that. However, comparing only the effect of considering and without considering the Soret effect, the difference is very small. The notable differences are seen at around 5 mm from the nozzle centre at 35 mm radial location, and at the nozzle centre at 75 mm radial location. This small difference however was not a result of the temperature changes as in the radiation study, but affected by the different profile of CO$_2$ mass fraction (please refer Fig. 8 in [125]) where at 35 mm, underestimation of the CO$_2$ when considering Soret effect gives a larger Rayleigh ratio value compare to the one at 75 mm, overestimation of the CO$_2$ giving produced otherwise, due to the larger cross section compared to other species. By contrast, H$_2$ was hugely changed when the different model was used, but it does not affect the Rayleigh ratio much as its Rayleigh cross-section is small.

There are two main conclusions that can be depicted from this laminar oxy-fuel flame study. First, the choice of radiation model is very important in comparing the Rayleigh ratio signal as this radiation model gives a huge impact to the temperature value while only little changes in the species distribution profile. As shown in equation (4.6), the Rayleigh ratio is calculated by considering the temperature and also the species distribution. Different radiation was shown to hugely change the temperature profile, however less influence in the species distribution changes. Obvious Rayleigh ratio changes when employing different radiation model drew a conclusion that the temperature dominated the changing of Rayleigh ratio in different radiation models. Second, the Soret effect influences only a small temperature change. However, as expected it gives a notable difference in the species profiles especially when considering a high diffusivity molecule such as H$_2$. This change in species profile however gives only modest changes in the Rayleigh ratio, which are mostly dominated by the change in larger cross section species such as CO$_2$, but less pronounced to the change of overall Rayleigh ratio.
Figure 4-5: Comparison of experimental and numerical simulation with and without Soret effect (a) experimental (b) P1 simulation with Soret effect (c) P1 simulation without Soret effect. Comparison of experimental and simulation with and without Soret effect along a line at (d) 35 mm and (e) 75 mm.
4.4.2 Comparison of experimental and numerical Rayleigh signals in turbulent flame

Figure 4-6: Single shot comparison (for qualitative illustration) of turbulent flame at Re=18,000, left (experiment) and right (simulation). Bottom (23 mm – 48 mm), middle (48 mm-73 mm) and top (98 mm – 123 mm).
For the turbulent flame, the comparisons are made in terms of mean and RMS values gained from the experiment and the numerical simulation for 1000 and 15000 shots respectively. The data were collected at three different windows locations, at 23-48 mm, 48-73 mm and 98-123 mm, which at every measurement location, a line at a specific axial location (35 mm, 55 mm and 110 mm) as shown in the single shot illustration in Figure 4-6 is extracted and discussed in this section. Overall, the flame structure captured by numerical simulation agrees well with the experiment at those three locations. At the lowest measurement window, the flame is thin and it gets broader with the increasing of measurement height. This can clearly be seen in Figure 4-7 where the mean and variance are presented. By referring to the variance Rayleigh ratio (right), the variance of the numerical calculation is always overestimated where the overestimation is calculated to be up to 5 %, while the Rayleigh ratio is underestimated for about 7 %. The variance is also seen to get thicker and broaden from the lowest to the highest measurement.

The detailed quantification is made by plotting single line data at three different axial locations namely 35 mm, 55 mm, and 110 mm as shown in Figure 4-8 (left). The mean Rayleigh ratio as discussed previously is seen to be underestimated by the simulation at 35 mm and 55 mm and agrees well with the experiment at 110 mm. The Rayleigh ratio is also seen thinner compared to the experimental data at 35 mm, however get broaden further downstream and agrees well at 110 mm. This comes from the little influence of the differential diffusion, which consideration of Le=1 gives a thinner flame as shown in laminar flame analysis in Figure 4-5. This trend was also discussed by Meier et al. [56] which concluded that at the upstream location, the thermochemical state of the flame cannot be described satisfactorily with only one Lewis number (the simulation in this work only considers Le=1), while at downstream location, Le=1 should instead give a good agreement. Similarly, the peaks of the RMS are also seen underestimated at upstream locations, and improving further downstream.

To further analyse the recorded and simulated signals, PDFs of Rayleigh ratio are plotted in Figure 4-9 which they are based on different criteria, one using data from locations near the peak in the mean profile (left), while the other from strongest fluctuations (right), both based on Figure 4-8 of the simulation values. In the left column, the PDF values get broader as the height is increased. On the other hand, the highest PDF is found to be at the
upstream measurement location and decreases downstream. Comparing the shape of the PDF, the numerical prediction shifts towards the lower Rayleigh ratio value for the lowest measurement location, and improves as it gets higher, and agrees well with the experimental data further downstream. On the other hand, when comparing at the strongest fluctuation locations (right column), the numerical gives a very good estimation to the experimental data, and agree well at all measurement locations.

The numerically calculated Rayleigh ratio were successfully compared and analysed statistically in detail in the turbulent flame and the qualitative comparison agreed well with the experimental data. Quantitatively, the analysis which was based on mean and RMS revealed that there is a small deviation in upstream region due to the little influence of the differential diffusion. However, this improved when the signals compared further downstream. Further investigation with PDFs revealed the same result.
Figure 4-7: Comparison of the (a) mean and (b) variance of the Rayleigh ratio from experiment and numerical simulation. The experiment is always on the left, while numerical on the right.
Figure 4-8: Comparison of the experimental and numerical data: mean (left) and RMS Rayleigh ratio (right)
Figure 4-9: PDF of Rayleigh ratio: using data at the location of the peak in the mean profile (left) and data from locations which show strongest fluctuations (right).
4.4.3 Comparison of normal and inverted flame structure using Rayleigh and OH-PLIF signals

A joint measurement of several signals could be used to further validate the numerically simulated signals to achieve high confidence in flame modelling especially in turbulent flames. Realising the need to provide a reliable model, an effort has been made to further analyse and validate the current model by looking at Rayleigh ratio as well as OH-PLIF signals of both experimentally recorded and numerically calculated data. By employing this joint measurement, the correlation of the Rayleigh ratio and the OH-PLIF signals can be analysed. In this joint measurement experiment, a turbulent inverted flame experiment has also been performed in addition to the normal turbulent flame to extend the study that have been performed by Wu et al. [22] and Takagi et al. [23] in laminar flames. In this work, a comparison of these two types of flame configurations is made focusing on the structure influenced by the turbulence.

Both of the normal and inverted flames were operated at the same jet and co-flow velocities, 30 m/s and 0.2 m/s respectively. This comparison by maintaining the jet and co-flow velocities in the inverted and normal flames has been done previously by Takagi et al. [23] in simple H₂-air laminar flames. However, a limited number of study in the turbulent inverted flame as well as in oxy-fuel flame have initiated this investigation.

Figure 4-10 shows the comparison of the Rayleigh ratio and OH-PLIF signals of normal and inverted flames at 67 mm from the jet nozzle. As seen in the section 4.4.2, the numerical simulation underestimates NDF Rayleigh ratio at upstream location, at the fuel rich region. However, in the IDF configuration, the structure and the thickness of the numerical calculated Rayleigh ratio agrees well, but shifted to nozzle centre. The RMS value is from both numerical and experimental agrees well in IDF compared to the NDF. Comparing the IDF and NDF specifically, the IDF is seen to be thinner than the NDF, as what have been reported by [23]. Takagi et al. [23] however reported that in laminar H₂/N₂-air flame, the temperature in IDF is much higher compared to NDF due to a stronger differential diffusion resulting from the differences in the H₂ ratio. Contradicting this earlier report, in this flame there is only little difference of Rayleigh ratio between the NDF and IDF meaning the same temperature was observed for both configurations for this mixture.
For the OH-PLIF comparison, the signals are normalised to the maximum value at the particular line. Structurally and looking at the measurement signal in terms of the OH-PLIF thickness, both of the flame configuration exhibit the same trend, and well captured by the numerical simulation, especially for the IDF. However, the numerical calculation for both signal lean towards the nozzle centre. As explained earlier, this comes from the little influence of the differential diffusion, which consideration of Le=1 gives a thinner flame.

Figure 4-11: Comparison of the PDF at different radial positions of the Rayleigh ratio RMS peak, OH-PLIF mean peak and OH-PLIF RMS peak at height of 67 mm from nozzle Experimental data (top) and numerical data (bottom) for IDF flame.
The overestimation of the OH-PLIF RMS signal is also seen in NDF flame.

To assess the ability of numerical simulation to capture the dynamics and flame structure, different locations that exhibit different structure have been chosen as marked in Figure 4-10, and the joint PDFs of IDF flame were made as shown in Figure 4-11. The first position is in the fuel lean region where the Rayleigh ratio and OH-PLIF increased almost linearly. The second column represents the high reading exhibited by both signals, and the third column is in the fuel rich region where the Rayleigh ratio is most probable at lower OH-PLIF ratio. The agreement between the experimental and numerical result can be considered good, even though some visible difference can be seen.

4.5 Uncertainty analysis

In this chapter, a comparison was drawn between the signal collected from the experiment and numerical calculation. For the experimental signals, minor post-processing was made, only to clean the data of stray light and normalise the flame signal by oxidiser signal. This normalisation also corrected for the laser sheet non-uniformity.

As discussed earlier, the Rayleigh ratio is calculated using equation (4.5):

\[
R_R = \frac{S_R^{\text{avg} \ OX}}{S_R^{\text{flame}}} = \frac{T}{T_{\text{ox}}} \left( \frac{\sum_{i=1}^{n_{\text{OX}}} X_i \sigma_{Ri}}{\sum_{j=1}^{n_{\text{flame}}} X_j \sigma_{Rj}} \right) = \frac{T}{T_{\text{ox}}} \frac{\sigma_{OX}}{\sigma_{\text{flame}}}
\]

As the temperature was not derived from the collected signal, much uncertainty is avoided. The uncertainty involved in the Rayleigh ratio is then reduced to temperature dependence of the Rayleigh cross-section, laser pulse energy fluctuation and laser sheet profile. Rayleigh cross-section was reported to be temperature-dependent by Sutton et al. [132] by up to 2 % in CH₄-air flame. However, Fuest et al. [63] reported in their DME flame variation only up to 1 %. In this flame, a linear relationship between the cross section and species temperature-dependence as reported in [132] was used and calculated using numerically-calculated species as a function of reaction progress variable. At the highest temperature, the maximum difference of applying the temperature dependent cross-section was found to be 2 %. The other possible uncertainty stems from laser fluctuations. In this experiment, energy fluctuation was not corrected as the variation from one shot to another was below 3 %.
(This was measured using an energy meter.) Finally, to test the uniformity of the laser profile, the intensity fluctuation in every oxidiser image in a region of uniform gas composition was compared row by row from top to bottom of the image. 30 Columns were first averaged to reduce the effect of shot noise. The stability of the laser beam and the profile uniformity were confirmed to be acceptable; normalisation of every image to the average oxidiser image would suffice given the variation from image to image at any given location was less than 3% in Rayleigh ratio measurement. The combination of the error from these sources can then be determined and for a single-shot signal, the combined maximum error in Rayleigh ratio measurement was 4.7%.

4.6 Conclusion

A comparison of Rayleigh ratio and OH-PLIF signals from experimental measurement and numerical simulation were performed in oxy-fuel flames. A tailored fuel and oxidiser combination (fuel: 17.5% CH₄, 40% CO₂, 42.5% H₂, oxidiser: 68% O₂, 32% CO₂) were chosen in the fuel selection process to ensure the temperature of interest (>2400 K) is achieved and at the same time making sure the soot formation is at its minimum level to avoid measurement errors. The stray light reflected from measurement surfaces was carefully managed to reduce at its minimum level, and the remaining was corrected using oxidiser and helium signals. The Rayleigh signal was normalised to the oxidiser signal to give “Rayleigh ratio” which eliminated some experimental constants and enabled numerical comparison. The OH-PLIF signal was collected under linear laser excitation conditions and normalised (at each given height) before comparison for a semi-quantitative measurement. The numerical Rayleigh ratio and OH-PLIF signals were estimated from the simulated temperature and species mole fraction.

The first comparison was made in laminar flame, where the kinetic mechanism sensitivity study was performed. However, only little influence to the considered fuel/oxidiser mixture was revealed, and GRI 3.0 mechanism was employed. The radiation effect was also investigated using P1 and OTM approaches. Temperature change was highly seen to change rather than the species composition, and highly influenced the absolute signal values with the OTM underestimates the Rayleigh ratio signals, and overestimated by adiabatic. The P1 approach was found to be closest to the measurement result. The Soret effect was also
analysed, with the spatial redistribution of H$_2$ and CO$_2$ being the most obvious effect, along with a moderate temperature change, overall resulting in a small but noticeable change to the Rayleigh ratio.

In the turbulent flame investigation, statistical comparison of the mean and variance or RMS as well as pdfs between experiment and numerical simulation was performed. The mean Rayleigh ratio was seen to be underestimated by the simulation and seen thinner compared to the experimental data at 35 mm and 55 mm above nozzle. However, further downstream at 110 mm the numerically calculated Rayleigh ratio agreed well with the experiment. Several pixels representing 1) the location of the peak of the mean profile and 2) the region of strongest fluctuation were picked to compare the distribution of the Rayleigh ratio values. Furthest upstream, the numerical pdf deviated towards small Rayleigh ratio values, improving further downstream. The “strongest fluctuation” pdf in contrast was captured well by the simulation in all measurement locations.

An extended study for the joint measurement of Rayleigh ratio and OH-PLIF was also performed to further validate the numerical simulation. Two flame configurations, namely the normal diffusion flame and inverted diffusion flame were compared using the same jet and co-flow velocities. Again, at upstream region, numerical calculation underestimates NDF Rayleigh ratio, in the fuel rich region while in IDF configuration the numerically-calculated Rayleigh ratio agrees well, but shifted slightly towards nozzle centre. Comparing both IDF and NDF flames, there was not much difference between the structure as well as the Rayleigh ratio. The latter indicates that there is no significant peak temperature difference between these two configurations.

From this investigation, it was shown that it is feasible to validate the numerical simulation to the experimental data by comparing the Rayleigh ratio and OH-PLIF signals for the purpose of improving the numerical model. The two-dimensional temperature measurement ability of the Rayleigh technique coupled with a flame front marker from the OH-PLIF signal was used to compare the overall structure (including snapshot comparison/visualisation), and provided the ability to extract any information at any measurement location for comparison.
We have demonstrated that the direct comparison of signals works well for oxy-fuel combustion. This approach of direct comparison for model validation is also not dependent upon a certain fuel/oxidiser mixture as the “traditional” Rayleigh technique would be. This has the dual benefit of enabling a wider range of fuel and oxidiser combinations to be tested as well as enhancing the confidence that a model can be validated accurately.
5 Simultaneous mixture fraction, temperature, OH- and CH₂O-PLIF measurement

5.1 Introduction

In this chapter, a further investigation of oxy-fuel flames is described. The investigation is focussed on the effect of varying O₂ in oxidiser as well as Reynolds number at elevated temperature in the non-premixed jet burner. Simultaneous measurement of temperature, OH and CH₂O is the main objective, in order to understand the correlation between these quantities in oxygen enhanced oxy-fuel flames. The data collected from this investigation would be useful in validating computational modelling as well as extending knowledge of oxy-fuel turbulent combustion and laser diagnostics.

In the previous chapter, a direct signal comparison was shown to successfully work to validate a numerical model. However, often the physical quantity (e.g. temperature) is solely of interest and numerical model validation is a secondary concern. Hence, this chapter will continue to discuss oxy-fuel flame structure, and explore the feasibility of employing an alternative Rayleigh scattering technique that enables temperature to be accurately determined in oxygen enhanced oxy-fuel flames.

The investigation involves a joint measurement of two-dimensional mixture fraction and temperature by using polarised/ depolarised Rayleigh scattering, and OH/CH₂O species using laser induced fluorescence technique. This polarisation/depolarisation technique is preferred over ‘traditional’ Rayleigh as the latter is vulnerable to change in relative Rayleigh cross section which gives an estimated temperature error of about 20-40 % in this fuel and oxidiser combination. The polarisation state of scattered light in ‘traditional’ Rayleigh scattering experiments has typically been ignored. The relative signal difference between measured signal in these two polarisation orientations as a result of a carefully-chosen combination of fuel and oxidiser can be exploited to determine the flame mixture fraction, and hence the temperature could be easily and accurately extracted. CH₂O (formaldehyde) is important in combustion since it acts as a reaction precursor and as a preheat zone indicator. OH exists in the oxidation layer and post-flame zone. In this chapter, these two
species are simultaneously probed in order to reveal further understanding of the evolution of the oxy-fuel combustion process.

5.2 Methodology

5.2.1 Fuel and oxidiser selection

In this section, the selection process of the fuel and oxidiser mixtures that were used in this work is presented. This covers analysis on the expected polarisation and depolarisation signal difference, as well as the sensitivity analysis of the Rayleigh signal difference as a result of using different strain rates and transport models, which are all based on

\[ \text{Figure 5-1: Species and temperature profiles of fuel: 55\% H}_2, 45\% \text{ CH}_4, \text{ oxidiser: 50\% O}_2, 50\% \text{ CO}_2 \text{ calculated using CHEMKIN employing opposed jet laminar flame, (a) } a=100 \text{ s}^{-1} \text{ and } 1000 \text{ s}^{-1}, \text{ equal diffusivity (b) } a=100 \text{ s}^{-1} \text{ and } 1000 \text{ s}^{-1}, \text{ multi-component. Temperature, normalised polarised and depolarised Rayleigh cross-section at different strain rate } (a=100 \text{ s}^{-1} – 1000 \text{ s}^{-1}) \text{ using (c) equal diffusivity (d) multi-component transport.} \]
numerically calculated result.

The determination of mixture fraction using reduced formulation to form a conserved scalar based on work demonstrated by Stårner et al. [133] from simultaneous measurement of two quantities through different fuel concentration (Raman or LIF)/Rayleigh imaging techniques were shown by Frank et al. [134]. It was found out that there is a drawback associated with each of the method studied which are weak Raman signal in fuel Raman scattering, fuel pyrolysis in the fuel-rich side of flame using fuel fluorescence, and dependencies of fuel tracer to the temperature in molecular tagging fluorescence technique. However, an approach introduced by Frank et al. [50, 74] through the use of polarisation and depolarisation Rayleigh signal could overcome the limitations and drawbacks of the techniques mentioned earlier. Furthermore, the nature of Rayleigh technique itself that has a high SNR and coupled with a suitable fuel and oxidiser combination was shown as a success.

The same aspects of flames in terms of keeping the soot formation at a minimum level (to successfully apply the Rayleigh technique) and making sure they can be operated at high Reynolds number were considered to determine the right fuel/oxidiser. These were explained in detail in section 4.2.2. After careful consideration, taking into account the flame properties as well as their suitability to be investigated employing Rayleigh difference technique (this will be explained next), a decision was made to employ fuel/oxidiser mixtures of fuel: 55 % H₂, 45 % CH₄, oxidiser 1: 50 % O₂, 50 % CO₂ and oxidiser 2: 55 % O₂, 45 % CO₂ in this work. 50 % O₂ flame and 55 % O₂ flame will be used for the oxidiser 1 and oxidiser 2 respectively hereafter. Note that 50 % and 55 % of O₂ (by volume) in the oxidiser stream were chosen based on two reasons: 1) to study the effect of varying O₂, but keeping the adiabatic temperature at around 2500 K and 2) further addition of O₂ above 55 % (and at the same time reduction of CO₂) in the oxidiser stream was observed enhancing soot formation at downstream flame region. Numerical calculations of laminar flamelets for these fuel and oxidiser combinations were performed using CHEMKIN-PRO employing opposed jet reactor considering GRI 3.0 mechanism approach [126] and the calculated results for the 50 % O₂ flame are presented in Figure 5-1. Similar trend was observed for the 55 % O₂ flame and thus is not presented here. Both the equal diffusivity and multi-component transport models were simulated covering the expected range of strain rate of
a=100 s⁻¹ to 1000 s⁻¹ where the global strain rate of the opposed jet flames was defined following [135] as

\[ a = \frac{2v_{ox}}{L} \left( 1 + \frac{v_f \sqrt{\rho_f}}{v_{ox} \sqrt{\rho_{ox}}} \right) \]  

(5.1)

where \( v \) and \( \rho \) represents velocity and density, while \( ox \) and \( f \) indicates oxidiser and fuel stream respectively. \( L \) is the separation distance between the two jet nozzles.

Figure 5-1 (a) and (b) represents the simulated species and temperature for the equal diffusivity and multi-component models at \( a=100 \) s⁻¹ and 1000 s⁻¹ respectively. Altogether, 18 species were considered. The stoichiometric mixture fraction was simulated to be 0.0853. The main species were plotted individually, while the species that contribute to mole fraction in a range of 0.01 to 0.1 (O, H, and OH) were summed up and plotted as ‘main intermediates’. The OH constitutes of about 60 %, while the other two represent 20 % of the overall value. The remaining species (C, CH₃, CH₂O, C₂H₄, C₂H₆ and C₃H₈) were also added up and presented as ‘others’. Both of the diffusion models are seen to produce the same trend, but the significant difference can be seen in some species such as H₂, CH₄ and CO₂. The peak of H₂O is also seen to be lower in equal diffusivity model compared to the multi-component, at about 0.44 and 0.48 respectively. Contrary to the H₂O, CO is produced more in equal diffusivity model compared to multi-component, though not much different giving the peak value of 0.22 and 0.21 respectively. Reducing the strain rate will result in increasing of these two species (not shown here). The same trend is also seen for the ‘main intermediates’ and ‘others’ and the later was seen to reduce to half of its value with the decreasing of 1000 s⁻¹ to 100 s⁻¹.

The ‘main intermediates’ species have previously been neglected such as in Raman/Rayleigh measurement in dimethyl ether (DME) and lower temperature oxy-fuel flames performed by Fuest et al. [63] and Sevault et al. [57] respectively. The contribution of this ‘main intermediates’ in high temperature oxy-fuel flame, however, cannot be neglected as it represents of more than 5 % of mole fraction as shown in Figure 5-1 (a) and (b). In term of polarised Rayleigh signal, this weight up to 4.5 % of the overall signal. For the ‘others’ species, even though the concentration is only about 0.8 %, but due to a bigger Rayleigh
cross-section for some of the species (C\textsubscript{2}H\textsubscript{4}, C\textsubscript{2}H\textsubscript{6} and C\textsubscript{3}H\textsubscript{8}), this contributes to almost 3 % of the overall polarised Rayleigh signal. The simulated temperature and calculated polarisation and depolarisation Rayleigh cross-section using the simulated species and temperature at different strain rates (a=100 s\textsuperscript{-1} – 1000 s\textsuperscript{-1}) are shown in Figure 5-1 (c) and (d) for equal diffusivity and multi-component respectively. The different diffusion models almost do not affect the peak temperature values if a comparison is made at the same strain rate. However, decreasing the strain rate (from 1000 s\textsuperscript{-1} to 100 s\textsuperscript{-1}) will result in temperature increase, from 2380 K to 2532 K for equal diffusivity, and 2370 K to 2550 K for the multi-component model. The peak locations for both models were also seen shifted to a leaner side of mixture fraction. For the polarised and depolarised Rayleigh cross-section, the highest difference between the two strain rates was also found at the leaner side, at mixture fraction of around 0.064. The increase of strain rate decreases the effective Rayleigh cross-section at mixture fraction of 0 – 0.1, due to the decreasing concentration of CO\textsubscript{2} and H\textsubscript{2}O which have large Rayleigh cross-section and replaced by a higher concentration of CO (through dissociation of CO\textsubscript{2} at high temperature) and ‘others’ which have smaller Rayleigh cross-section. Beyond that, the concentration of CO\textsubscript{2} is getting higher again (in a=1000 s\textsuperscript{-1} compared to a=100 s\textsuperscript{-1}) and so the Rayleigh cross-section. Looking at the overall trend of the polarised Rayleigh effective cross-section (from mixture fraction 0 to 1), the variation is about 40 % as a result of a bigger effective Rayleigh cross-section at mixture fraction.

Figure 5-2: (a) predicted polarised and depolarised Rayleigh ratio, signal difference and signal difference using non-linear combination of polarised and depolarised Rayleigh signal calculated at strain rate of 800 s\textsuperscript{-1}, (b) non-linear combination Rayleigh signals at different strain rates (100 s\textsuperscript{-1} – 1000 s\textsuperscript{-1} (top) and the percentage difference of the Rayleigh signals (bottom).
fraction below than 0.15 and almost constant beyond that, again mainly because of the CO₂ concentration.

The correct combination of isotropic (such as CH₄, He, Ar) and anisotropic (such as CO₂, O₂, N₂) molecules in the fuel and oxidiser mixtures is critical to ensure a good contrast between the polarised and depolarised signals and this discriminate between fuel rich and fuel lean regions. Isotropic molecules scatter only parallel light, while the anisotropic molecules scatter both parallel and perpendicular lights to the incoming polarised laser source. These two different scattered light orientations were imaged using two different cameras and with the knowledge of the Rayleigh cross-section of each species for both orientations, the effective Rayleigh cross-sections and hence the polarised and depolarised Rayleigh signals can be calculated. Predictions of the polarised and depolarised Rayleigh ratio of 50 % O₂ flame calculated at a strain rate 800 s⁻¹ are shown in Figure 5-2 (a). It can be seen that the polarised Rayleigh ratio is multivalued, and this clearly cannot be used alone to predict the mixture fraction. However, with the help of the depolarised signal, where the fuel combination was chosen to have a higher depolarised signal in the lean while having a small signal in the rich region, the difference between these two signals can be used to determine the mixture fraction. However, a simple signal difference has a very shallow gradient (and thus low sensitivity) especially in the mixture fraction range of 0 – 0.2, which includes the stoichiometric value. This problem was overcome by applying a non-linear combination to both depolarised $R^\text{dep}_R$ and polarised $R^\text{pol}_R$ Rayleigh ratio signals, where all constants were chosen on trial-and-error basis, read as

$$S_{nl} = R^\text{pol}_R^{0.5} - \left(2.5R^\text{dep}_R\right)^{0.5} + 1 \quad (5.2)$$

This non-linear combination of the two Rayleigh signals significantly improves sensitivity, especially at the higher temperature region, between mixture fraction 0.05 – 0.1. Quantitatively, the signal difference (over mixture fraction range f=0 to 0.2) was improved from just 5 % to about 48 %.

A wide range of global strain rate of the fuel/ oxidiser combination was calculated to analyse its impact on the Rayleigh signals. Figure 5-2 (b) shows the non-linear combination Rayleigh signal at strain rates of 100 s⁻¹ to 1000 s⁻¹, for both diffusion models. The multi-component signal is predicted to have a higher signal compared to the equal diffusivity
model. However, the difference between the two extremes (multi-component, $a=100 \, s^{-1}$ and equal diffusivity, $a=1000 \, s^{-1}$), is below than $\pm 2 \%$, which this translates to $5 \%$ error in mixture fraction. However, this error is mainly confined to the fuel rich region, where the predicted temperature based on the CHEMINK calculation is $<1200 \, K$. In this region, the error is expected to be about $\pm 60 \, K$, while at the peak temperature this falls to $\pm 40 \, K$. Though the correct strain rate is required to get an accurate temperature reading, based on the sensitivity analysis it does not have too much impact on the fuel/ oxidiser mixtures discussed in this chapter.

5.2.2 Experimental setup

![Diagram of experimental setup]

*Figure 5-3: Experiment setup for simultaneous mixture fraction, temperature, OH- and CH$_2$O-PLIF measurement*

The diagram in Figure 5-3 shows the experiment arrangement that was used in this chapter. This measurement system is an extended system from the one that was explained in section 4.2.3, hence only a brief description is given here as this section is only intended to explain
the additional optics and light collection equipment used. The Rayleigh scattering was induced by a combination of three spatially overlapped frequency-doubled Nd:YAG laser heads, generating about 1.5 J/shot. Another cavity from the multi-channels Thales laser was frequency triplet generating third harmonic wavelength at 355 nm from its fundamental of 1064 nm. The energy generated was around 270 mJ and was used to excite the CH$_2$O transition of $A^2A_1 \leftarrow X^1A^1_0 pQ$. Even though this excitation scheme is rather weak compared to another transitions such as at 339 nm [136, 137] and 353 nm [110, 138], the high energy available from the frequency tripling of Nd:YAG laser definitely compensates this drawback. Furthermore, the other transitions require an employment of another dye laser, which would further complicate the setup and increase the cost of the experiment. This high laser intensity is also being the most reliable method to saturate polycyclic aromatic hydrocarbons (PAH) [108]. For the OH-PLIF, the same Sirah PrecisionScan dye laser which use Rhodamine 6G solution dissolved in ethanol as described in section 4.2.3 was used, pumped by a 250 mJ frequency-doubled Nd:YAG laser generating 5 mJ/pulse to excite $Q_1(8)$ line of the $A^2\Sigma^+-X^2\Pi(1,0)$ transition at around 283 nm. The 355 nm and 283 nm beams were first overlapped using a dichroic mirror, and these two beams were further overlapped to 532 nm beam using another dichroic mirror that reflects the 283 nm and 355 nm while transmitting the 532 nm. These overlapped beams were then transformed into light sheets with a height of 8 mm for all sheets with the help of iris and thickness of 70 μm, 75 μm and 80 μm for the 532 nm, 355 nm, and 283 nm respectively using a combination of cylindrical and spherical lenses. These sheets thicknesses were measured by rotating the positive cylindrical lens to 90 degrees, and Rayleigh images air were recorded for the 532 nm while the 355 nm 283 nm sheets were imaged using fluorescence signal produced by biacetyl and acetone in quartz cell respectively. The lens combination that transforming the beams into light sheets was first adjusted to ensure that the narrowest waist of the 532 nm sheet was at the centre of the burner nozzle. The other two beams (283 nm and 355 nm) waisted at a different location due to chromatic aberration and so a telescope (combination of a positive and negative spherical lens) was placed in each to adjust this shifting.

A combination of polarisation and depolarisation Rayleigh signals was used to determine the mixture fraction and temperature in this experiment. The polarisation Rayleigh signal was collected using un-intensified interline transfer CCD camera (LaVision Imager Intense),
coupled with a 105 mm f/2.0 Nikon lens while the depolarisation Rayleigh signal was collected using an ICCD camera (LaVision imager intense plus IRO) coupled with 85 mm f/1.2 Mitakon Speedmaster lens. Both of the CCD cameras has 1280 x 1024 pixels, with a pixel size of 6.45 μm and the quantum efficiency of about 60 %. A GaAsP photocathode IRO was used for the depolarisation Rayleigh imaging (with a quantum efficiency of around 45 % at 532 nm, about five times higher than the typical S20 photocathode). A narrow band 532 nm interference filter (Δλ_{FWHM} = 3nm), and a B+W Käsemann HTC Polarizing Filter were fitted to both Rayleigh camera lenses to optimise the suppression of stray light and ensure collection of only horizontally and vertically polarised Rayleigh signal for the depolarisation and polarisation Rayleigh respectively. The gate time for the polarisation Rayleigh was set at 1 μs while the depolarisation Rayleigh camera was gated at 230 ns.

The OH-PLIF signal at \( \Delta \Sigma^+ \rightarrow \Delta \Pi \) (1,1) and (0,0) transitions peaking at around 308 nm was collected using ICCD camera (LaVision imager intense plus IRO) with a 100 mm Halle AR-coated UV lens (f/2.0) fitted with colour glass filters (1mm thick Schott UG5 with 90 % transmission and WG305 with transmission of more than 95 % at 308 nm) to block the elastic scattering, and background luminosity was reduced by setting up the gate at 200 ns. The P46 phosphor, S20 photocathode LaVision IRO used in this experiment has a quantum efficiency of about 14 % at 308 nm. The choice of this excitation scheme has already been discussed in (section 3.1.3.1 and section 4.2.3) and will not be discussed here.

For the CH\(_2\)O fluorescence signal, an ICCD camera built from a LaVision Imager Intense and P43 phosphor, S20 photocathode IRO was used where it has a quantum efficiency of around 15 % in the fluorescence detection range (400 nm – 500 nm). The camera was equipped with a 105 mm (f/2.8) Sigma EX Macro lens fitted with long and short pass filter of 400 nm and 500 nm respectively to reduce interferences from Raman and further reduce the remaining PAH signal. The gate time was set to be 350 ns which was long enough to collect all the CH\(_2\)O fluorescence and short enough to block all other interferences.

The cameras arrangement was made as in the Figure 5-3 to ensure that the depolarisation Rayleigh camera can be equipped with the 85 mm (f/1.2) to collect the light at its maximum, as with an arrangement of two cameras at each side would require a longer focal lens and a dichroic mirror which will reduce the light transmitted to the camera sensor. Two dichroic
mirrors were used to separate all three wavelengths, which the first one was used to reflect 300 - 320 nm for the OH-PLIF and transmit the 400 – 700 nm. The second dichroic mirror was then used to reflect 400 nm – 500 nm for the CH₃O signal and transmit the wavelength above 500 nm for the Rayleigh signal collections.

All the images from four cameras were aligned and distortion correction was also applied using LaVision DaVis software, with sub-pixel accuracy. Energy monitor was employed in every laser line to record all the energy in single-shot basis. This energy data were used to correct the energy fluctuation for all images. The fluctuation for the 532 nm and 355 nm beams were in the order of 3 % (Thales laser is equipped with temperature control units). The 283 nm beam, however, showed about 10 % fluctuation. This was attributed to the fact that the BBO crystal responsible for frequency doubling was not equipped with any temperature control.

5.2.3 Post processing

<table>
<thead>
<tr>
<th>Flame</th>
<th>Composition</th>
<th>Jet Reynolds</th>
<th>Co-flow velocity</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>Fuel: 55 % H₂, 45 % CH₄</td>
<td>7000</td>
<td></td>
</tr>
<tr>
<td>A2</td>
<td>Oxidiser: 50 % O₂, 50 % CO₂</td>
<td>12000</td>
<td></td>
</tr>
<tr>
<td>A3</td>
<td></td>
<td>18000</td>
<td></td>
</tr>
<tr>
<td>B1</td>
<td>Fuel: 55 % H₂, 45 % CH₄</td>
<td>7000</td>
<td>0.3 m/s</td>
</tr>
<tr>
<td>B2</td>
<td>Oxidiser: 55 % O₂, 45 % CO₂</td>
<td>12000</td>
<td></td>
</tr>
<tr>
<td>B3</td>
<td></td>
<td>18000</td>
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</tr>
</tbody>
</table>

*Table 5-1: Experimental conditions for simultaneous mixture fraction, temperature, OH- and CH₃O-PLIF measurement*

The experimental condition for this experiment is shown in Table 5-1. The two different flame compositions with three different jet Reynolds numbers each was tested. The co-flow velocity, however, was kept constant. In each test case, a set of images of flame (1000), oxidiser (700), argon (700), helium (700), acetone (500), biacetyl (500) and dark current (500) were recorded. The step to post-process all the images will be explained next.
5.2.3.1 Polarisation and depolarisation post processing

The post-processing for the polarisation and depolarisation Rayleigh were done first by correcting the energy fluctuation using the energy data collected during the experiment. The stray light contribution for the polarised Rayleigh images was calculated as explained in section 4.2.4, using equation (4.4).

For the depolarised signals, other factors have to be taken into account such as the extinction of the polariser (i.e. unwanted transmission) and also the orientation of that polariser relative to the laser polarisation, even though a great care was taken to ensure only depolarised Rayleigh could pass the filter. The latter was checked by rotating the polariser fitted onto the depolarised camera lens until the scattered light signal collected from argon gas was minimised (as an isotropic molecule, it should produce no depolarisation signal). The remaining signals that get into the camera are then only the stray light and leakage signal from the polarised Rayleigh. The leakage signal, \( S_{\text{leak}} \) is dependent on the intensity of the polarised molecule Rayleigh signal, \( S_{R}^{\text{Pol}} \) which can be expressed as

\[
S_{\text{leak}} = C_{l} S_{R}^{\text{Pol}}
\]  

(5.3)

where \( C_{l} \) is the leakage constant. This leakage constant was determined by imaging helium and argon gases and can be expressed using

\[
C_{l} = \frac{S_{\text{Dep,Ar}}^{\text{Raw}} - S_{\text{Dep,He}}^{\text{Raw}}}{S_{\text{Pol,Ar}}^{\text{Raw}} - S_{\text{Pol,He}}^{\text{Raw}}}
\]

(5.4)

where \( S_{\text{Dep,Ar}}^{\text{Raw}}, S_{\text{Pol,Ar}}^{\text{Raw}}, S_{\text{Dep,He}}^{\text{Raw}}, S_{\text{Pol,He}}^{\text{Raw}} \) are the raw depolarised and polarised signal of argon and helium respectively. These two gases were chosen as they produce zero depolarisation signal, and helium produces small, while argon relatively large polarised signal. Note that the depolarised helium image can be used directly to calculate the stray light contribution as 1) helium gas produces zero depolarised signal and 2) helium produces a weak polarised signal which when attenuated by the leakage factor would be negligible. In order to complete this analysis it was also assumed that the polarised signal from helium is negligible compared to argon, which is reasonable because the scattering cross section is 66 times smaller.
After the initial post-processing that corrects energy fluctuations, particle cleaning, stray light and leakage, a typical resulting image is as shown in Figure 5-4 (a), the left representing polarised signal and the right depolarised. The non-uniformity of the laser sheet is pronounced in the oxidiser region (on the right side of the images) as the oxidiser has a larger Rayleigh cross-section compared to the fuel, hence produces a higher intensity of scattered light. The next step was normalising the flame images to the oxidiser using an average oxidiser image using equation (5.5) and (5.6) for polarised, $R^\text{Pol}_R$ and depolarised, $R^\text{Dep}_R$ Rayleigh ratio respectively. This also cancels out the non-uniformity of the laser sheet as shown in Figure 5-4 (b).

$$R^\text{Pol}_R = \frac{S^\text{Pol}}{S^\text{avg Ox,Pol}} = \frac{(S_{\text{raw}} - S_{\text{stray}} - S_{\text{dark}})_{\text{Pol}}}{(S_{\text{raw}} - S_{\text{stray}} - S_{\text{dark}})_{\text{Ox,Pol}}}$$ (5.5)

$$R^\text{Dep}_R = \frac{S^\text{Dep}}{S^\text{avg Ox,Dep}} = \frac{(S_{\text{raw}} - S_{\text{He}} - S_{\text{dark}})_{\text{Dep}}}{(S_{\text{raw}} - S_{\text{He}} - S_{\text{dark}})_{\text{Ox,Dep}}}$$ (5.6)

The signal difference of polarised and depolarised Rayleigh ratio as shown in Figure 5-4 (c) was then calculated. As explained in section 5.2.1, a non-linear combination of these two signals, written as $S_{nl} = R^\text{Pol}_R^{0.5} - \left( (2.5)R^\text{Dep}_R \right)^{0.5} + 1$ was used to increase the slope or sensitivity at the fuel lean region. It was experimentally impossible to identify the mole fraction of each scattering species, however, following the reasoning given earlier in this chapter (see e.g. Figure 5-2b), a simulation run in an opposed jet laminar flame in CHEMKIN-PRO employing GRI-MECH at a strain rate of 800 s$^{-1}$ was performed to discover the evolution of $S_{nl, \text{ sim}}$ and Rayleigh cross-section ratio, $\sigma_{\text{Ray, ratio}} = \frac{\sigma_{\text{Ox}}}{\sigma_{\text{flame}}}$ as a function of mixture fraction. These relationships were used to produce images of experimental mixture fraction and Rayleigh cross-section as shown in Figure 5-4 (d) and (e). Finally, with the knowledge of the oxidiser temperature, $T_{\text{Ox}}$ and with a constant pressure, the flame temperature $T_{\text{flame}}$ can be expressed as

$$T_{\text{flame}} = \frac{T_{\text{Ox}}}{R^\text{Pol}_R} \left[ \frac{1}{\sigma_{\text{Ray, ratio}}} \right]$$ (5.7)
A typical instantaneous single shot temperature is shown in Figure 5-4 (f), calculated using equation (5.7) taking into account the Rayleigh cross section variation. The variation of the Rayleigh cross section is large (from reactants to products). If the variation is not considered and the temperature is calculated assuming the cross section is constant, this will result in
overestimation in the fuel and in the high temperature region and could result in as much as 40% error giving a temperature up to 3500 K as shown in Figure 5-4 (g).

An analysis has been done to understand in detail the sensitivity of experimentally measured temperature to different strain rates. The analysis involves a wide range of strain rate that is expected to happen in this type of flame from 400 s\(^{-1}\) to 1000 s\(^{-1}\), for both equal and multi-component diffusivity models as shown in Figure 5-5. The analysis was performed in the burner radial space, and mixture fraction space, where the \(\Delta T\) is the difference of temperature to the average temperature indicating the overall uncertainty of using different strain rate is less than 3%. The trend of the temperature that is shown here same with the trend of the signal difference or \(S_{nl}\) that was shown in Figure 5-2 (a) where a high signal difference occurs at the fuel rich and fuel lean (indicated by temperature here), but meet somewhere at around mixture fraction of 0.1.

5.2.3.2 OH-PLIF post processing

For the OH-PLIF processing, the same initial procedure was done by correcting the energy fluctuation to the image using the recorded energy. The image was then normalised to an average acetone image to correct the laser sheet non-uniformity. A semi-quantification is of interest and the OH concentration was calculated considering the LIF linear regime using equation (4.7) in section 4.3. The semi-quantification requires the knowledge of main species and temperature involves in the combustion process so that the quenching rate correction to the raw image can be done. The ability of the polarisation and depolarisation
Rayleigh explained in the previous section that enables the determination of mixture fraction, which in turn makes a proper estimation of the main species involved possible has helped the quenching correction to be made. The species composition that was used here was calculated using CHEMKIN-PRO, at 800 s⁻¹ strain rate as explained in section 5.2.1. The quenching rate, $Q_{21}$ was calculated using

$$Q_{21} = \sum_i N_i \sigma_i v_i$$  \hspace{1cm} (5.8)

where $N_i$ denotes as the number density of collisional partner, the collisional cross-section is denoted by $\sigma_i$ and $v_i$ is the relative velocity of the collisional partner and the excited molecule. The cross section of every species involving in the quenching is dependent on the temperature, and this was calculated using [61] equation (5.9) below, before quenching rate coefficient for every species can be determined.

$$\sigma_q = \sigma_{q\infty} \exp \left( \frac{\epsilon}{RT} \right)$$  \hspace{1cm} (5.9)

The sensitivity analysis of the OH concentration using different strain rate (the same analysis as done to the temperature) was also performed, giving the difference of less than 5 % to the final value.

### 5.2.3.3 CH₂O post processing

For the CH₂O processing, after the energy correction and normalisation of the CH₂O signal to the biacetyl fluorescence to correct the non-uniformity of the laser sheet, another correction, to minimise the CH₄ Raman signal in the images were done. The methane Raman bands peak were shown by Harrington and Smyth [108] appeared at around 398 nm and 424 nm. In this experiment with the use of dichroic mirror that only reflect the wavelength more than 400 nm, and a long filter used on the camera lens that transmits only 400 nm-500 nm, it was believed that the only CH₄ Raman that gets into the camera was the band at 424 nm.

The knowledge of mixture fraction has once again been very helpful in correcting the CH₂O signal that was contaminated with CH₄. The mole fraction of CH₄ conditioned on mixture fraction can easily be determined using the CHEMKIN simulation, and hence this was used to determine the fraction of the CH₄ that needs to be eliminated. The elimination process
was performed by averaging the CH$_2$O raw signals and finding a location (10 x 10 pixels) that has a mixture fraction of more than 0.98 (indicating pure fuel and no CH$_2$O, threshold chosen to account for sensor noise) as marked in red in Figure 5-6 (a). These pixels were then averaged, and the value used as a baseline for the correction at mixture fraction of 1 for all CH$_2$O images in this experiment for consistency. At all other locations, a relative CH$_4$ Raman signal was determined and correction was applied based on the local mixture fraction and thus mole fraction of CH$_4$. Figure 5-6(b) and (c) shows the corrected CH$_2$O signal after the CH$_4$ elimination, and a comparison between the uncorrected CH$_2$O and corrected CH$_2$O normalised average signal. This has improved the signal, but most importantly the elimination process does not change the structure of the CH$_2$O signal as the corrected and uncorrected traces overlap between 0.005 m to 0.015 m from the nozzle centre (fuel lean region). Therefore, the correction only eliminates the crosstalk in the fuel rich region.

CH$_2$O fluorescence processing also uses the same principle as the OH where a linear fluorescence model was used to calculate the CH$_2$O concentration. The signal, however, can
only be semi-quantitatively calculated due to insufficient quenching data. The evaluation of the $Q_{21}$ was done following the assumption made by [62] where it was assumed to vary between $Q_{21} \propto T^{-0.5}$ and $Q_{21} \propto T^{-1}$. The Boltzmann fraction, $f_b$ was calculated following the expression suggested by Dimitris et al. [114] considering the vibrational and rotational contribution to the population fraction while neglecting the population of electronically excited states which read as

$$f_b = \left(1 - e^{-1680.5/T}\right) \left(\frac{40.1969e^{-740/T}}{1 + \frac{0.134}{T} + \frac{0.037}{T^2}T^{3/2}}\right)$$  \quad (5.10)$$

The choice of either $T^{-0.5}$ or $T^{-1}$ for $Q_{21}$ in this type of flame has been done by processing the CH$_2$O signal and plotting it as shown in Figure 5-7. Figure 5-7 also compares the raw CH$_2$O signal as well as predicted signal calculate using CHEMKIN. This broader CH$_2$O profile has also been shown by Gordon et al. [71] where the comparison was made in Bunsen flame. In this work however, with the knowledge of the mixture fraction, the experimentally measured CH$_2$O signals can be compared easily to the calculated values from CHEMKIN. It is shown from this figure that the peak of the $Q_{21} \propto T^{-0.5}$ and $Q_{21} \propto T^{-1}$ shifted a bit to about mixture fraction of 0.2, while the simulated CH$_2$O peaking at around 0.16. Though the
experimental measured is broader than the simulated profile, the overall trend shows an acceptable agreement. The $Q_{z1} \propto T^{-1}$ was chosen for the post-processing value in this work as it is marginally better than the $Q_{z1} \propto T^{-0.5}$ being closest to the simulation value which is most clear between mixture fraction 0.2 – 0.6.

5.3 Result and discussion

5.3.1 Instantaneous, mean and RMS of temperature, OH and CH$_2$O signals analysis.

Figure 5-8: Selection of instantaneous triplet images (left) of temperature, OH and CH$_2$O at different Reynolds number: Re=7000 (bottom), Re=12000 (middle) and Re=18000 (top) for 55% O$_2$ flame. On the right is the mean (red lines) and RMS (black lines) profile of temperature, OH and CH$_2$O at 25 mm axial location for different oxygen concentration in co-flow.
Figure 5-8 (left) shows some selected instantaneous images of temperature, OH and CH_2O of 55% O_2 flame at different Reynolds numbers at axial location of 25 mm. Each of the images has a size of 8 mm x 30 mm and the nozzle centreline is marked with a white dashed line. It should be noted that every triplet image in this section is displayed in the order as shown in Figure 5-8 (left), temperature (top), OH (middle) and CH_2O (bottom). The structure of 50 % O_2 flame at this location exhibits a similar trend and thus is not shown here.

At this region, the laminarisation effect is seen taking place especially for the lower Reynolds number due to the increase of viscosity as a result of temperature increased of the gases. The local Reynolds number is hence reduced and so this effect is produced. This laminarisation effect at the region near to the nozzle has also been reported by Bergmann et. al [46]. This laminarisation effect acts as a shield avoiding any cold oxidiser entrainment and penetration into the reaction zone, which reducing the possibility of localised extinction. This effect however is reduced, which can be seen in a higher jet velocity especially at Re=18000 where the turbulent-like transition from the laminar-like shape is observed. The OH however remain intact and there is no evidence of localise extinction occurred in this region in both of the flames at all Reynolds number through observing the OH images. Observing the OH alone may not enough to conclude there is absence of localise extinction as the fluorescence lifetime of OH is much longer and might have been diffused to covering the small extinction regions, and indeed one may say the CH_2O is seen to penetrate into the reaction zone by observing the ‘purple’ colour dots in the CH_2O images the Figure 5-8 (left), however this signal is too low, which might be come from the CH_2O system noise. Localised extinction will be further discussed in section 5.3.4.

The thickness of the flame (visualised using OH and temperature images) at a low Reynolds number is seen thicker and decreasing with increasing of the Reynolds number. This phenomenon can be explained by the longer time of the molecules in lower Reynolds number to diffuse in the radial direction. When the Reynolds number in increased, this diffusion time is reduced and hence reducing the thickness of the flame.

The other behaviour that can be seen from the instantaneous images is the high intensity of the CH_2O at a higher Reynolds number compared to the lower one. This observation is consistent with the laminar opposed jet flame calculation using CHEMKIN software, which
the CH$_2$O is simulated to have a higher concentration at a higher strain rate. The increase of the CH$_2$O has shown by Medwell et al. [73] which due to the partial premixing through the enhanced mixing at the high Reynolds number. The other possible reason is most probably the CH$_2$O has been consumed in a lower Reynolds number as it has a longer time for the chemical reaction to take place compared to the one at high Reynolds number.

Figure 5-8 (right) shows the mean and RMS temperature, OH and CH$_2$O profiles for both of the flames. This mean and RMS were generated using a strip of 1 mm (20 pixels in axial direction) height, and only the right half (i.e. from the nozzle centreline to the right) was considered from every image. The latter was to ensure any beam steering and absorption effect in the LIF measurement is avoided. No beam steering is observed in the Rayleigh signal, however some absorption effect can be seen especially in OH-PLIF images as the signal on the right side of the images is always higher compared to the left.

As expected, the mean temperature of the 55 % O$_2$ is higher than those 50 % O$_2$ as presented in the Figure 5-8. The calculated adiabatic temperature of 55 % O$_2$ is 2670 K while the 50 % O$_2$ is 2600 K. The temperature measurement data were analysed and all single shot temperature measurement is below the adiabatic temperature, indicating good data reliability. It can be seen that increasing the Reynolds number causes a small drop in the mean temperature which is consistent with the calculation performed in CHEMKIN, suggesting a lower strain rate (represented by Re=7000) has a higher temperature compared with the higher one (Re=18000), which also has been noted by Bergmann et al. [46].

The decreasing of Reynolds number is also seen to increases the thickness of the OH reaction zone outwards from the nozzle centre, with all the OH mean profile at different Reynolds number shifted to that direction. This reaction zone thickness represented by OH will be discussed further in section 5.3.3. The CH$_2$O profile is hardly changed, while the temperature profile is seen maintain their inner position besides the broadening on the outer layer. As discussed earlier in the single shot images, the mean CH$_2$O at Re=7000 is lower compared with the higher Reynolds number, with almost 20 % different for 55 % O$_2$ flame.
Comparing the two flames structures, the OH mean is seen insensitive to the change of oxygen concentration displaying a very similar OH peak concentration. The increase of OH concentration is generally related to increase in temperature, which is not quite the observed result even though from the CHEMKIN calculation the peak difference should only be around 15 %. The peak RMS for both of the temperature and OH in 50 % O₂ is also higher, suggesting there is enhance of mixing. This is also supported by the reduction of CH₂O concentration which is believed has been consumed generating that high OH.

Figure 5-9: Selection of instantaneous triplets images (left) of temperature, OH and CH₂O at different Reynolds number: Re=7000 (bottom), Re=12000 (middle) and Re=18000 (top) for 55 % oxygen flame. On the right is the mean (red lines) and RMS (black lines) profile of temperature, OH and CH₂O at 50 mm axial location for different oxygen concentration in co-flow.
Figure 5-9 (left) shows the instantaneous images of 55 % O$_2$ flame at 50 mm height above the nozzle. The effect of laminarisation still appears for the Re=7000, and the CH$_2$O is seen still at a minimum level compared to the higher intensity of CH$_2$O generated at higher Reynolds number. For the Re=12000 the turbulence has seen giving some impact to the flame structure and the flame has started to wrinkle, and the CH$_2$O has started to clearly appear in the fuel rich region as a result of the fuel decomposition, following the shape of the OH as shown in the middle image.

By observing the OH images there is no evidence of localised extinction at the two low (Re=7000) and middle (Re=12000) Reynolds numbers, even though occasionally a thin reaction zone appears. At Re=18000 however, breaks in the OH have started to appear indicating localised extinction is happening. From the observation of the overall images, these breaks account for 2 % of the overall images at this Reynolds number at 50 % oxygen, while in 55 % oxygen this happens at less than 1 % from the 700 images. The higher oxygen concentration has therefore started to play its role, as fewer breaks are seen (compared to the 50 % O$_2$ flame) in the OH indicating that the flame is experiencing a higher reaction rate. This higher reaction rate is the result of a reduced chemical time scale. There is also an increase in viscosity which helps to maintain an intact OH “ribbon” and avoids penetration of cold flow into the reaction zone. Figure 5-9 (top left) showing that the OH reaction zone has ruptured allowing the fuel to get premixed with the oxidiser producing the product of preheat zone i.e. CH$_2$O and this intermediate species will be consumed and reignited in a less turbulent intensity region downstream [139].

This localised extinction has caused the temperature drop as shown in the single shot temperature image in the Figure 5-9 (top left). The mean temperature for both flames at this 50 mm region is also seen to be lower than at 25 mm as shown in Figure 5-9 (top right). This is opposed to the flame stretch theory as when the axial position is increased, the velocity and velocity gradient is decreased and so the flame stretch. The 50 mm hence should have a higher temperature than those at 25 mm. To make a fair comparison, avoiding the temperature drop caused by localised extinction at Re=18000 and the strong effect of diffusivity at Re=7000, the flame at Re=12000 lends itself to comparison. The temperature difference between the 25 mm and 50 mm axial locations is 96 K and 112 K for 50 % and 55 % flames respectively. This higher temperature behaviour near the jet was also
observed by W. Meier et. al [55] in non-premixed H₂-air turbulent flames, where the temperature at x/D=5 was measured 100 K more than the adiabatic temperature.

This was reported due to the effect of unequal rates of mass diffusion of the different species and heat conduction. The effect of differential diffusion were also reported by Bergmann et al. and Meier et al. [46, 56] and they concluded the thermochemical state of flames near the flame base is complex and usually consists of a combination of Lewis numbers as well as strain rates.
Both of the flames at this axial location show that they get broadened with the increasing of axial location. The mean and RMS of OH and CH\(_2\)O show the same trend as at 25 mm axial location, displaying very similar mean peak value of OH and a lower mean CH\(_2\)O value for 50 % O\(_2\) flame as compared to 55 % O\(_2\) flame. As expected, the mean temperature at 55 % is shown to be higher than the 50 % O\(_2\).

At 75 mm axial location, the temperature, OH and CH\(_2\)O profiles shown broader than those at 50 mm and 25 mm. Opposed to the mean temperature at lower axial locations, the flame at higher Reynolds number at this location is seen broader than the lower, for both 50 and 55 % O\(_2\) flames with the latter being more obvious. The RMS temperature for the 55 % flame is also seen to be higher than the 50 % flame at this location. There is also an increase of mean OH for 50 % flame for Re=12000 and Re=18000 as well as the RMS values albeit not much compared to the other radial locations.

The trend of CH\(_2\)O for the 50 % is seen to decrease with increasing axial locations. However an opposite trend is observed in 55 % flame, which showing a higher CH\(_2\)O at 75 mm. Based on the high RMS temperature, as well as a high RMS CH\(_2\)O, this most probably as a result of enhanced turbulence mixing between the fuel and oxidiser, and at the same time increase the premixed reaction. One thing to note, the temperature for Re=7000 in Figure 5-10 (top right) is shown lower than those at higher Reynolds number. The existence of soot is believed to produce such result, as further downstream a low local Reynolds number as well as low momentum increase the residence time which apparently enhance the generation of the soot. Rayleigh images contaminated by soot particles can easily be detected as the soot particles produce high Rayleigh signal. In post-processing stage, soot particle detection algorithm was used to detect such signal in every Rayleigh image. Any image detected with soot particle was then rejected. The existence of this soot and PAH has also affected the CH\(_2\)O signal giving the mean value three fold compared to the other CH\(_2\)O from other Reynolds number and thus is not shown here. The temperature, OH and CH\(_2\)O for Re=7000 at this location is hence not considered in this thesis. The temperature and OH that is shown in Figure 5-10 (top and middle right) is only for the purpose to illustrate how one can detect when there is an existence of contamination in the recorded signals.
Figure 5-10 (left) shows the selected single shot images for the temperature, OH and CH₂O at 75mm height above nozzle for 55 % O₂ flame for different Reynolds numbers. The turbulence has now shown reducing the laminarisation behaviour of Re=7000 compared to the other radial locations. For the Re=12000, the OH reaction zone has sometime become thinner even though the probability of it to gets broader as shown in the mean profile is higher. There is however no evidence of localise extinction to happen at this Reynolds number for both flames, again by observing the OH signal. The enhanced turbulence and the generation of premixed reaction as explained earlier have also resulted in a broader CH₂O region at this axial location. For the Re=18000, the localised extinction is seen to happen in both of the flames as at the 50 mm axial locations. There is an increase of localise extinction in the 50 % O₂ flame giving the occurrence of this event of about 4 % while in 55 % O₂ flame it is observed that this event occurs less than 1 % from 750 images.

Figure 5-11: Selected single shot triplet images of temperature, OH and CH₂O of 50 % O₂ flame at Re=18000 at 75 mm radial position
In addition to that, the generation of CH$_2$O pockets has also been observed to exist at this location for Re=18000 in both flames as shown in Figure 5-11. This is the evidence of enhanced mixing, and a result of the localise extinction that enables the premixing of the fuel and oxidiser which initiate the premixed flames through the penetration of the oxidiser into the fuel region. The re-ignition from the premixing process is now explained. One important point to note that the re-ignition process that will be explained with the aid of the images as stated previously are not from the ‘true’ sequence order from the recorded images, instead a selection of images were done to represent this event. The ‘true’ sequence would require a high speed system to resolve, but in this experiment the images were taken at only 10 Hz. This low speed system however is more than enough to understand the structure of the high temperature oxy-fuel flame, as it being one of the objectives of this experiment.

The re-ignition process is proposed to occur in four steps, as shown in Figure 5-11 (a) – (d). Firstly, the localise extinction due to the rupture of the reaction zone has to happen enabling the oxidiser to get into the fuel zone, or vice versa and thus promote the premixing between these two as shown in Figure 5-11 (a). Then this premixing between the fuel and oxidiser will cause fuel decomposition, and thus the CH$_2$O pockets are generated as shown in Figure 5-11 (b). This CH$_2$O pockets are possibly to get re-ignited at downstream region, at a region where the local Reynolds number is low and has a longer residence time for the chemical reaction to take place. The re-ignition also requires enough temperature for the pocket CH$_2$O to get re-ignited. If the reaction process is successful, this will then increase the local temperature and hence the OH is generated. Figure 5-11 (c) shows where the CH$_2$O is in the process of re-ignition, and the OH has started to be produced as well as there is a rise in local temperature. This re-ignition process will continue until the all CH$_2$O is consumed and the peak of OH is generated as shown in the Figure 5-11 (d). The CH$_2$O is believed to have just been consumed due to evidence from the CH$_2$O ring surrounding the produced OH peak. It should be noted that in Figure 5-11 (d), there are 2 important regions in the CH$_2$O image: 1) CH$_2$O ring as evidence the re-ignition process has taken place (top of the image), 2) another CH$_2$O pocket which is believed to be consumed with enough local temperature to get it re-ignited.
5.3.2 Further analysis on pocket OH and CH$_2$O

![Figure 5-12: Example of localised extinction from 50 % O$_2$ flame at 50 mm radial position. The single shot images of temperature, OH and CH$_2$O (top), joint PDF of temperature and OH (middle) and CH$_2$O and temperature (bottom).](image)

In this sub-section, further analysis on the re-ignition process based on selected single shot images is presented. This analysis is done using the joint PDF of OH and temperature as well as the CH$_2$O and temperature to understand how each of these quantities reacted with the process. All the analysis in this section is done using the single shot images from 50 % O$_2$.
flame at 50 mm axial position, where all the presented images were manually selected (i.e. visualise every image). For each process, the analysis were done from several events and they produced almost similar joint PDF structure and so only two of the selected events for each process are presented here. The data of the joint PDF is taken only from the dotted box marked in grey.
As explained earlier in the previous section, the re-ignition process is initiated from the premixing of the fuel and oxidiser. For this process to take place, the oxidiser has to get into the fuel region or vice versa through the rupturing of the reaction zone. The entrainment from the cold oxidiser exists and is shown dropping the temperature in that region, where the highest temperature is read at around 2300 K and peaking at below 1500 K as shown in Figure 5-14.
Figure 5-12. The OH is of course at its minimum level, and the CH$_2$O is seen to start building up though the lower intensity of CH$_2$O is still dominating the sampled region. From both of the CH$_2$O joint PDFs, the CH$_2$O intensity is shown to linearly increase with temperature. This CH$_2$O pocket will be then consumed if the surrounding temperature is high enough to re-ignite it.

Figure 5-15: Example final stage of the re-ignition process from 50 % O$_2$ flame at 50 mm radial position. The single shot images of temperature, OH and CH$_2$O (top), joint PDF of temperature and OH (middle) and CH$_2$O and temperature (bottom).
Figure 5-13 shows some images that have a higher probability of the CH₂O compared to the one that shown earlier in Figure 5-12. This indicates that the fuel decomposition process continues to take place and this process is further enhanced by the high temperature. The high surrounding temperature will definitely increase the chance of the CH₂O pocket to get re-ignited, providing that the chemical reaction rate is high enough. The temperature is seen sometimes to almost achieve the peak temperature of the flame. The consumption of the CH₃O at high temperature will produce the OH as the intermediate in the post-flame region. This can be seen in the Figure 5-13 (bottom) which the increase of OH is the evidence that the re-ignition process has now started.

As the process continues, the production of the CH₂O has arrived at its maximum, and at the same time, the production of OH is continued and this increase the OH as presented in Figure 5-14. The probability of temperature is also seen peaking and shifted to a higher temperature region. Figure 5-15 shows the final stage of the re-ignition process. Here, the sampling area (box size) was increased to cover the ‘consumed’ CH₂O area as well as its ring. The joint PDF of OH is shown to increase to its maximum, and this is clearly seen in the single-shot images where high intensity OH is present in the pocket, while the CH₂O has been almost consumed and the probability peaks at a lower quantity as shown in Figure 5-15 (bottom). Note that the OH concentration distribution peaks at lower quantity as well, however this is due to the enlargement of the sampling area.

5.3.3 Reaction zone thicknesses of OH

The thicknesses of the reaction zone were measured using the OH images. This was achieved by first setting the minimum threshold of the processed OH signals, and any pixels below this threshold level were set as 0 while the opposite to 1. Then, the edges of the OH islands were detected. Next, the centrelines of the islands were determined and the thickness was calculated from the centreline to the edge of each side. Note that the thicknesses were collected only from the right side (from nozzle centre to the right of the images) of all the 750 OH images, and the PDF of each case was generated as shown in Figure 5-16. Overall, the OH thicknesses from the two different O₂ concentrations were distributed very similar. However, it can be noticed that at Re=7000 especially at the lowest measurement window, the PDF of the 55 % O₂ were shifted towards thicker reaction zone
albeit the profile shape is maintained similarly to the 50 % O₂ flame. This is believed comes from the laminarisation effect together with the increasing of the reaction rate in the 55 % O₂ flame as explained earlier. At the lowest position, the laminarisation effect shields the reaction zone from the entrainment or penetration of the cold air, and this strong shielding at the same time maintains the thickness of OH layers. When it gets further, the turbulent and cold air entrainment highly affects the OH thickness, resulting in a broader range of thickness.

![Figure 5-16: PDF of OH thicknesses for both flames at Re=7000, Re=12000 and Re=18000 at different radial location](image)

The average thicknesses of each flame, at each Reynolds number at different axial locations are presented in Table 5-2. The result indicates that the OH thickness is increase with the increase of axial location. This is expected and consistent with the result reported by Kelman et al. [139]. Secondly, the increase of the oxygen also increases the OH thickness through the increase of the reaction rate. Thirdly, the increasing of the Reynolds number has a
noticeable effect in the decreasing of layer thickness at lower radial location. At the highest recorded position however, the trend does not follow. This is due to the domination of the turbulent flow field when it gets further downstream.

<table>
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<th>x, mm</th>
<th>25 mm</th>
<th>50 mm</th>
<th>75 mm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>50 % O₂</td>
<td>55 % O₂</td>
<td>50 % O₂</td>
</tr>
<tr>
<td>Re=7000</td>
<td>1.440 mm</td>
<td>1.590 mm</td>
<td>1.797 mm</td>
</tr>
<tr>
<td>Re=12000</td>
<td>1.102 mm</td>
<td>1.175 mm</td>
<td>1.669 mm</td>
</tr>
<tr>
<td>Re=18000</td>
<td>1.013 mm</td>
<td>1.038 mm</td>
<td>1.590 mm</td>
</tr>
</tbody>
</table>

Table 5-2: Mean layer thickness of OH reaction zone

### 5.3.4 Scatter plot of temperature and localised extinction analysis

![Figure 5-17: Scatter plot of temperature at x/D=5 (25 mm) and x/D=10 (50 mm) for 50 % O₂ flame at Re=12000 and Re=18000. The stoichiometric mixture fraction, f_{st}, is marked with grey dotted vertical line.](image)
Analysis of the correlation between temperature and mixture fraction often requires a complex experimental setup. However, using the polarised/depolarised Rayleigh technique, this can be achieved. In this section, the correlation between the temperature and mixture fraction at different axial positions, as well as different Reynolds number are analysed. The analysis is done by comparing measured temperature to different set of calculated temperature data using different strain rates and diffusion model.

Figure 5-17 shows the correlation of temperature and mixture fraction for Re=12000 and Re=18000 at two different near-field positions at x/D=5 (or 25 mm) and x/D=10 (or 50 mm). It should be noted that only a line of pixel from each image from 750 images were plotted. The stoichiometric mixture fraction is plotted with the grey dashed line. To allow the comparison between the recorded data and numerical calculation, the plotted data is overlaid with calculation results at different strain rate as well as Le=1 and Le≠1. Thus, a range of strain rate using different diffusion models at 15 s$^{-1}$ to 2500 s$^{-1}$ were calculated and

![Graphs showing temperature and mixture fraction correlation](image)

*Figure 5-18: PDF of temperature conditioned on narrow band mixture fraction ($\Delta=0.02$) centred at highest global average temperature for both flames at Re=12000 and Re=18000 at different axial position*
will be used as comparison. It should be noted that some scattered plots (forming almost a straight line) at \(x/D = 10\), at \(\text{Re}=12000\) (near the stoichiometric line) and at \(\text{Re}=18000\) (at the top-left of the figure) are most probably generated by the system noise. The small fraction of the noise (comes from several images) would not affect any analysis presented in this section, however.

By looking at the fuel lean side of both Reynolds number at \(x/D=5\), the calculated temperature is lower than the measured temperature. The strained laminar flame calculated temperature at different strain rates and different Lewis number underestimates the temperature at that region. This type of fuel and oxidiser combination is seen not to change much at the very fuel lean side as all the plotted temperatures overlap on top each other at the very fuel lean region. Approaching the stoichiometric, the strain rate at 1000 s\(^{-1}\) with \(\text{Le}=1\) is seen to match both of the profiles. In the same region at \(x/D=10\) however, all the calculated strained rates match to the measured temperature, nicely sit in the middle of the measured temperature. The measured temperature profiles however are lower than strained calculated temperature when they reach the stoichiometric. It also can be seen that the higher temperature region for both flames at \(x/D = 5\) are shifted to the fuel rich side. This is in line with the result reported by Sevault et al. [57] and this behaviour has also been reported by Law et al. [140] which is due to the product dissociation and the direction of shifting was reported based on the peak heat release. From the study, they found out that the dissociation in H\(_2\)-air mixture is more towards the lean side, and so the peak heat release occurs on the rich side. This behaviour is seen to reduce with the increasing of the radial location. At the mixture fraction between 0.2 to 0.5, all the calculated strained temperatures are seen to be lower than the measured, though the \(\text{Re}=18000\) at \(x/D=10\) nearly match the calculation at \(a=1000\) s\(^{-1}\), \(\text{Le}=1\). The same behaviour was also observed by Meier et. Al. [55] and they reported this is due to the unequal rates of mass diffusion for the different species as well as heat conduction. From the mixture fraction of 0.5 to 1, both of the temperature at \(x/D=5\) match to the calculated strain of 1000 s\(^{-1}\), \(\text{Le}\neq 1\), however the temperature profiles at \(x/D=10\) match the same strain rate but in between the \(\text{Le}=1\) and \(\text{Le} \neq 1\). As can be seen from the calculated temperature, taking the \(a=1000\) s\(^{-1}\) as an example the deviation between the two diffusion model is pronounce near the stoichiometric and at the fuel rich region. The comparison presented here has shown the importance of
considering this effect as at the near field the Lewis number is somewhat complex and cannot be described alone.

From the scatter plot especially at Re=18000, it can be seen that there are a significant number of outlier points at relatively low temperature, indicating localised extinction from partially combusted gases. It shows that the Re=18000 at 25 mm has a higher localised extinction compared to other Reynolds number and at different position. However, from the OH images observation that was explained in Section 5.3.1, it was found that the OH layer breakage starts to happen at 50 mm, and there was no evidence that localise extinction happens at the lowest position. There are two possible explanations for this. First, the localised extinction captured by this scatter plot might come from small regions and could not be clearly seen using the OH images. Second, a longer fluorescence lifetime of the OH might have covering the localised extinction region after it gets diffused to that region.

Figure 5-18 shows the PDFs of temperature conditioned on a narrow band mixture fraction at different Reynolds number and axial positions. These PDFs were determined using a narrow band mixture fraction ($\Delta=0.02$) centred at the highest global average temperature following the method reported in Sevault et al. [57]. For example, based on the temperature vs mixture fraction profile shown in Figure 5-17, the mixture fraction at which the highest average temperature occurs is determined. A narrow band mixture fraction is then set and all the temperature samples that fall into this band are used to build the PDF in Figure 5-18. From the figure, the peak and PDF profile at axial location of 25 mm at Re=18000 is shown to decrease and shift to the left compared to the peak and profile at Re=12000. The decrease in the peak of the PDF indicates the temperature distribution gets broadened, while the shift to the left indicates the distribution tends to low temperatures. Hence it can be said that higher localised extinction is expected in the Re=18000 (25 mm, 50 % O2 case). In other flames, the difference between the two Reynolds numbers was not noticeable. At 50 mm, Re=18000 for both flames shows a small shift to lower temperatures which might come from oxidiser entrainment into the reaction region. One thing to note is that the strain rate would affect the flame temperature, as a higher strain rate produces a lower temperature.
Again, following the method presented by Sevault et al. [57], to further quantify localised extinction, a minimum threshold temperature value below which the mixture is believed partially or not combusted can be specified. Threshold values were set as 1500 K (for 50% O₂ flame) and 1600 K (for 55% O₂ flame). All the flames show a probability of localised extinction below 1%, with the Re=18000 at 25 mm 50 % O₂ flame the highest at 0.7 %. However, if the threshold value is increased to 1800 K and 1900 K respectively for the 50 % O₂ and 55 % O₂, higher local extinction is observed in the downstream region; both flames reach almost 4 % compared to a maximum around 2.3 % in the upstream region. This method is therefore very sensitive to the choice of the threshold value which might limit its ability to provide a quantitative assessment of the probability of localised extinction.

5.4 Uncertainty analysis

The uncertainties found in the Rayleigh measurement discussed in the Chapter 4 were generally related to the equipment (such as laser sheet uniformity and energy fluctuation), though the temperature dependence of the Rayleigh cross-section was also included. In this chapter, as temperature was derived from the Rayleigh signals, the uncertainty was indeed higher. The temperature dependence of the Rayleigh cross-section and the laser sheet uniformity are applicable here too, but laser pulse energy fluctuation was corrected (and will be in Chapter 6 and 7 too) and hence can be neglected.

The temperature measurement employed the polarised/ depolarised Rayleigh technique. The signal difference, \( S_{nl} \) between these two signals (polarised and depolarised Rayleigh) was used to estimate the mixture fraction and thus the per-pixel Rayleigh cross-section was determined based on looking up results from a numerical calculation. Estimation of the mixture fraction from the signal difference was made based on the calculated flame using a strain rate of 800 s⁻¹ and employing a multi-component diffusion model. Sensitivity analysis was performed by means of comparing the calculated prediction of the signal difference at different strain rates and using diffusion models as shown in Figure 5-2. The maximum uncertainty between the different strain rates to the one used here was 2 %, leading to an error in mixture fraction space of 5 %. Experimental data was used to analyse the sensitivity of strain rate and diffusion model choices on temperature (please refer Figure 5-5 ). It was shown that the highest temperature uncertainty is about 3%, mainly on the fuel rich and
fuel lean side, while keeping quite low near stoichiometric. Combining all these uncertainties in quadrature, the total uncertainty in the temperature measurement is estimated to be about 7%.

Knowledge of the temperature and mixture fraction from the polarised/depolarised Rayleigh experiment is very beneficial for Boltzmann fraction and quenching corrections in LIF signals. For the OH, temperature was used to correct the Boltzmann fraction variation while the experimentally-measured mixture fraction data was used to determine the colliding species’ concentrations from the numerical results. Sensitivity analysis on the strain rate used for the total quenching value was also calculated giving a maximum error of 5%. The total combined OH concentration error includes the beam non-uniformity is thus estimated to be around 10%. For the CH$_2$O, temperature was also used to correct the Boltzmann fraction and quenching. However there is no species-specific quenching data available, so a general quenching correction was made using the approach suggested by Paul and Najm [62]. The error in CH$_2$O concentration after quenching correction was specified to be 10% [114]. Combining this error with the temperature error, the overall error in CH$_2$O concentration is expected to be around 15%.

5.5 Conclusion

A joint measurement of mixture fraction, temperature, OH- and CH$_2$O-PLIF has been performed in non-premixed jet burner using oxy-fuel flames. For the mixture fraction and temperature measurement, polarisation and depolarisation Rayleigh technique was employed. The success of this technique relies mainly on the fuel/oxidiser combinations to being able determine the mixture fraction which is used for the temperature correction in the post-processing, as without the correction 20-40 % temperature error is expected in oxy-fuel flame. Careful analyses were performed and two fuel/oxidiser combinations were selected. The temperature and composition information from the polarised/depolarised Rayleigh was used to identify accurate quenching corrections for both OH and CH$_2$O LIF signals. The compositional information recovered from the polarised/depolarised technique also enabled a novel correction of the formaldehyde PLIF signal for suspected contamination from a Raman signal generated by methane.
Simultaneous data from the two different flames with varying oxygen content in oxidiser were acquired. For each flame, the Reynolds number was varied and the measurement was taken at three different axial positions. Overall the mean and RMS of the temperature, OH and CH$_2$O exhibits the same structure comparing at the same axial position. The laminarisation effect is seen to dominate at the x/D=5 especially for lowest Reynolds number (Re=7000), and the flame starts to get wrinkled when the Re or the measurement position is increased indicating the influence of turbulence. The reaction zone thickness measured from the OH signal has been shown to get broader with increase of axial position and thinner with increase of the Reynolds number. The addition of the O$_2$ in the oxidiser has also shown to increase the thickness albeit less significantly.

The joint diagnostics enabled physical insight and visualisation of the localised extinction and subsequent re-ignition processes, including identifying underlying correlations. In the first step, localised extinction enables premixing of the fuel and oxidiser and with enough temperature, there follows decomposition into CH$_2$O. The re-ignition process takes place at high temperature during which the CH$_2$O is consumed and OH produced.

Finally, an attempt was then made to quantify the observed localised extinction. From the two-dimensional OH images, localised extinction was observed at x/D=5 and x/D=10 but only at the highest Reynolds number, Re=18000. It was more frequent in the 50% O$_2$ flame compared to the 55% O$_2$ flame. However, following a previously published method Sevault et al. [57], localised extinction determined from a narrow band mixture fraction centred at the highest global temperature (i.e. using polarised/depolarised Rayleigh data alone) showed localised extinction to be most likely at x/D=5, Re=18000 in the 50 % O$_2$ oxidiser flame. This small discrepancy between the Rayleigh-based metric and the OH-PLIF assessment could be due to the finite lifetime of the OH radical which causes some spatial smearing, sealing small holes which are still detected by the temperature-only method.

This chapter has demonstrated the feasibility of recovering temperature in oxygen-enhanced oxy-fuel flames using polarised/ depolarised Rayleigh. The uncertainty analysis of this technique and the Rayleigh ratio technique in the Chapter 4 has shown that for validation purposes, Rayleigh ratio should be the best in terms of minimising experimental error, as the temperature inferred in this chapter requires several assumptions in terms of
strain rates, diffusion model etc. However, if the physical quantity (i.e. temperature) data is required, the technique explained in this chapter is more suitable. Care should be taken in choosing the strain rate for the Rayleigh cross-section determination when inferring the temperature data.
6 Analysis on the applicability of heat release represented by [OH][CH$_2$O] in non-premixed oxy-fuel flames

6.1 Introduction

The importance of heat release rate (HRR) in understanding the chemical conversion into thermal energy is crucial. The combustion noise and instability prediction in gas turbine requires the knowledge of HRR. This released energy is a product of wide range of reactions that take place in the combustion processes. In order to quantitatively determine the HRR, joint scalars measurement involving all species taking part in combustion has to be performed. The feasibility of such joint measurement is indeed not practical and impossible. In laminar flame configuration, Najm and Paul [68] performed a numerical and experimental study to understand the relationship between the HRR and other species that could be a marker for that HRR. The study reported that HCO profile is closely matched to that HRR. HCO signal however is very week due to a low concentration in the combustion process. In another work, Paul and Najm [62] demonstrated that the product of pixel by pixel multiplication of OH and CH$_2$O signals, [OH][CH$_2$O] could be used to represent the forward reaction of \( OH + CH_2O \to HCO + H_2O \). The resulting spatial and temporal correlation between the OH, CH$_2$O, HCO and the heat release was shown to be in agreement with the numerical calculation of laminar stoichiometric and rich flames in their study. A quantitative representation of the product of multiplication of the OH and CH$_2$O however should be done carefully, through the correction of the temperature dependence of the two LIF signals. The OH LIF signals could more easily be quantified as the collisional quenching [61] and the Boltzmann fractions data are available. The problem however lies on the CH$_2$O signal where the quenching data is not available. As discussed in Chapter 5, Dimitrios et al. [114] performed a study to understand the relation between the temperature and the CH$_2$O signal, and the quenching in CH$_2$O is proposed to be represented by \( Q_{21} \sim T^{-0.5} \) or \( Q_{21} \sim T^{-1} \).

Extensive studies have been performed to understand the applicability of this technique in premixed and non-premixed configurations, as well as in laminar and turbulent flames.
Some of the studies include the turbulent premixed flames in swirl burner performed by Bockle et al. [141] and Ayoola et al. [69] in CH4-air flame. In non-premixed flame, a study performed by Gordon et al. [142] shows a reliability of this technique to understand auto-ignition events. A recent study in turbulent spray flames has also been conducted by Yuan et al. [72] showing the applicability of this technique in n-heptane flames.

In this chapter, the applicability of the product [OH][CH2O] in representing the heat release in non-premixed oxy-fuel flames is explored. Analysis of the different behavior of heat release profiles in CH4-air laminar premixed, non-premixed and oxy-fuel non-premixed flames will be discussed. Further the correlation between the heat release/forward reaction rate to the temperature and the temperature gradient are discussed. Finally, the applicability of the reaction zone of [OH][CH2O] in predicting the local extinction is determined and compared to the temperature sampling method explained earlier in section 5.3.4.

6.2 Methodology

6.2.1 Temperature dependence of the LIF signals

In heat release rate imaging, there are two important temperature dependencies, 1) the LIF signal intensities, 2) the forward rate of reaction. However, these dependencies are usually neglected or assumed to have identical temperature dependence to reduce the measurement complexity. However this is not the case which has been shown by Mulla et al. [143] in their study of CH4/air premixed flames at equivalence ratio of 0.7-0.9. Frequently, the LIF signal from OH and CH2O is simply spatially multiplied pixel-by-pixel, [OH] x [CH2O] to represent the forward reaction rate (FRR) for $OH + CH_2O \rightarrow HCO + H_2O$.

A better representation of the rate of this reaction is however mathematically represented by

$$FRR = k(T)[OH][CH_2O]$$  
(6.1)
where \( k(T) \) is a specific rate constant and FRR is the forward reaction rate of \( OH + CH_2O \rightarrow HCO + H_2O \). The parameter \( k(T) \) is in Arrhenius\(^9\) form, and in this analysis, \( k(T) \) was determined from the laminar flame simulation using CHEMKIN. The dependencies of the LIF signals intensity towards the temperature by means of the collisional quenching and Boltzmann fraction population \( f(T) \) has been explained in the section 5.2.3. Though the heat release rate cannot be quantified due to the missing quenching data for \( CH_2O \) signals, the first step is to correct the temperature dependence. According to the literature, the quenching rate could vary by up to 15 % while \( k(T) \) varies by around 20 % over a temperature range of 1300-1900 K [143]. In this work, both the LIF signals and reaction rate are corrected for temperature.

### 6.2.2 Data reduction procedures

The post-processing of the LIF-OH and LIF-\( CH_2O \) signals has been explained in section 5.2.3. The post processed images of corrected collisional quenching and Boltzmann fraction were first averaged and the threshold value for each signal is set. The twenty highest values on every shot for all cases are then averaged (to reduce impact of shot noise biasing) to get the maximum intensity which then be used for the normalisation. Normalised images of the OH and \( CH_2O \) are then multiplied pixel by pixel and the product is then multiplied by the reaction rate constant \( k(T) \) (also using local temperature from simultaneous Rayleigh) to get the FRR images. Again, twenty points of maximum values from each reaction rate image are then collected and averaged for the normalisation.

For the temperature gradient images, the temperature images were first smoothed following the technique described by Frank and Kaiser [20] using inhomogeneous smoothing with an isotropic Gaussian kernel to reduce the unnecessary attenuation to the smallest length scale as well keeping an adequate noise suppression for the largest length scales. In temperature images, the largest length scales are those belongs to the high temperature and the smaller length scale are from the lower temperature. Applying this technique will result in varying the kernel size across the image; high temperature regions which have

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\(^9\) The Arrhenius equation is a formula that relates the chemical reaction rate to temperature. The equation is usually written as \( k = Ae^{-E_a/(RT)} \) where \( T \) is the temperature, \( A \) is the pre-exponential factor, \( E_a \) is the activation energy and \( R \) is the universal gas constant
lower signal to noise ratio will be smoothed with a larger kernel size while low temperature zones use a smaller kernel. The kernel size is calculated using

\[ \sigma_T = \sigma_0 \left( \frac{T}{T_0} \right)^\alpha \]  \hspace{1cm} (6.2)

Where \( \sigma_T \) is the temperature dependent kernel size, \( \sigma_0 \) is the reference kernel size, \( T \) is the local temperature, \( T_0 \) is the reference temperature and \( \alpha = 0.75 \) is the scaling exponent. The reference kernel size, \( \sigma_0 = 0.7 \) was chosen after performing an analysis using a homogeneous kernel filter, in order that the filtered image using this kernel size suffers minimal degradation. Finally, the temperature gradient, \( \nabla T \) was calculated by adding the two spatial components in quadrature:

\[ |\nabla T|^2 = \left( \frac{\partial T}{\partial x} \right)^2 + \left( \frac{\partial T}{\partial r} \right)^2 \] \hspace{1cm} (6.3)

### 6.3 Result and discussion

#### 6.3.1 The structure of the heat release profile

The applicability of the forward reaction rate, FRR, of \([OH][CH_2O]\) in \(CH_4\)-air premixed flames to represent the heat release was first shown by Paul and Najm [62]. A simple flame structure of premixed configuration enables the heat release layer to be determined easily. Opposed to this, in the non-premixed flames, the structure is more complex due to the diffusion. This results a different heat release profile, where in the premixed flames it consists only one zone of peak heat release, known as oxidation zone while in non-premixed flames, several zones (peaks) could exists namely fuel pyrolysis, primary oxidation and secondary oxidation zones as shown in Figure 6-1 (a) and (b). Hence, before heat release imaging is performed, especially in non-premixed flame, it is important to know by how much the product of \( k(T)[OH][CH_2O] \) underestimates/ overestimates (in term of contribution of HRR from secondary and fuel pyrolysis zones) the heat release rate, HRR. One thing to note that the heat release in premixed flame is plotted against position, while
in non-premixed the heat release is plotted against the mixture fraction. The premixed laminar flame was calculated using freely-propagating flame while the non-premixed laminar flame was calculated using opposed-flow diffusion flame, employing GRI Mech 3.0 [126] using CHEMKIN-PRO [119]. A multi-component diffusion model was used in the calculation. The reaction numbering (R10, R52 etc) is used following the reaction numbers in GRI Mech 3.0.

Figure 6-1: Comparison of total heat release rate and main reactions contributed to the total heat release in (a) premixed CH₄-air (b) non-premixed CH₄-air (c) non-premixed oxy-fuel flame of fuel: 55 % H₂, 45 % CH₄, oxidiser: 55 % O₂, 45 % CO₂ at strain rate of 2500 s⁻¹ using multi-component transport. A comparison of heat release and forward reaction rate of [OH][CH₂O] for fuel: 55 % H₂, 45% CH₄, oxidiser: 55 % O₂, 45 % CO₂ at different strain rates and Lewis number is shown in (d)

In both configurations, the major contributors to the total heat release in the oxidation zone come from the forward reactions:
In the non-premixed flames however, additional heat release comes from the reaction:

$$OH + CO \rightleftharpoons H + CO_2$$  \hspace{1cm} (R99)

The major contributor in reverse reaction for both configurations comes from R38, while in non-premixed, a reverse reaction of R166 is also to take place:

$$H + O_2 \rightleftharpoons O + OH$$  \hspace{1cm} (R38)

$$HCO + H_2O \rightleftharpoons H + CO + H_2O$$  \hspace{1cm} (R166)

In the secondary oxidation zone in non-premixed flames, other than small contribution from forward reaction of R84 and R99, and reverse reaction of R38, a forward reaction of R35 is also seen to contribute to the heat release:

$$H + O_2 + H_2O \rightleftharpoons HO_2 + H_2O$$  \hspace{1cm} (R35)

The pyrolysis zone in the non-premixed CH₄-air is seen contributed by the thermal decomposition of vinyl through the reverse reaction of

$$H + C_2H_2(+M) \rightleftharpoons C_2H_3(+M)$$  \hspace{1cm} (R71)

Figure 6-1 (c) shows the total heat release profile and the main contributors to the total heat release for non-premixed oxy-fuel flame of fuel: 55 % H₂, 45 % CH₄, oxidiser: 55 % O₂, 45 % CO₂ at strain rate of 2500 s⁻¹. The obvious difference comparing this flame to the CH₄-air flame is the broadening of the heat release profile as a result of wider reaction profiles. The region of the heat release profile in the oxy-fuel flame covers as early as $f=0.02$ to $f=0.5$, while in the CH₄-air flame, it started at around same mixture fraction, but ends at around
In the primary oxidation zone, most of the reactions behave the same way however, due to the high temperature the thermal decomposition of CO$_2$ into CO (the reverse of reaction R99) absorbs rather than releases heat. The primary to secondary peak ratio in the oxy-fuel heat release also reduces, due to the additional (secondary peak) forward reaction:

$$OH + HO_2 \leftrightarrow O_2 + H_2O$$  \hspace{1cm} (R287)

In the fuel pyrolysis zone which was observed in the non-premixed CH$_4$-air flame which was dominated by the decomposition of vinyl through the reverse reaction of R71, the same reaction is also taking place in the oxy-fuel non-premixed flame. However, due to a higher reaction rate of R52 as well as the addition of R158, the reverse of reaction R71 (negative heat release) is cancelled out and the result is a positive total heat release profile throughout the combustion process – from mixture fraction, $f=0$ to $f=1$.

$$O + C_2H_4 = OH + C_2H_3$$  \hspace{1cm} (R158)

Figure 6-1 (d) shows the total heat release rate (HRR) and forward reaction rate (FRR) for fuel: 55 % H$_2$, 45 % CH$_4$, oxidiser: 50 % O$_2$, 50 % CO$_2$ at strain rate of 400 s$^{-1}$ and 2500 s$^{-1}$ calculated using multi-component (Le≠1) and mixture average (Le=1) diffusion models. As explained in section 5.3.4, a single strain rate or diffusion model cannot capture the behaviour accurately. Thus comparing the calculated result from different strain rates with both models is required. The flames in this work are expected to operate between these two strain rates, and the figure shows that the HRR for a higher strain rate tends towards the fuel lean side while the HRR for a low strain rate towards the fuel rich side. The FRR behaves similarly. At a lower strain rate, the primary heat release zone is narrower compared to the higher strain rate, while the secondary zone region is much broader. The FRR covers only the primary oxidation zone, hence for a lower strain rate, FRR tends to underestimate the total heat release compared to higher strain rates. However, there is a good overlap between HRR and FRR in the primary oxidation zone at both strain rates, so
FRR can be used as a marker of the peak heat release location with increasing validity as the strain rate increases.

6.3.2 Comparison of simulated and measured OH, CH₂O and [OH][CH₂O] quantities

Before comparing the structure of the simulated OH, CH₂O and the FRR to the measured data, analysis was done to understand the effect of the dependency of the reaction rate on the temperature at different strain rates. As explained earlier in section 6.2.1, the LIF signals dependence on the temperature has been corrected. For the reaction rate dependency on the temperature, the parameter k(T) was calculated using CHEMKIN at different strain rates, to check whether this is another source of error that could possibly be introduced. Figure 6-2 (left) shows k(T) values at different strain rates. The two strain rate extremes give a maximum error of 2%. Note that there is an error of more than 20% if k(T) is assumed to be constant during analysis.

In a further analysis of Figure 6-1, normalised HRR is plotted against normalised FRR in Figure 6-2 (right) to understand the correlation between these two quantities at different strain rates. The black dashed line represents where all the data should be if the HRR and FRR were to agree with each other perfectly. Note that the peak heat release rate always correlates well with peak FRR. At a lower heat release rate, the FRR at strain rate 400 s⁻¹ and Le=1 is seen to underestimate the heat release under fuel lean conditions (see Figure 6-1 (c)). Changing diffusion model, strain rate 400 s⁻¹ and Le=1 gives reduced underestimation of
the lean heat release, but slight overestimation in the fuel rich region due to negative heat released linked to fuel pyrolysis and thermal decomposition of CO$_2$.

Figure 6-3 shows a comparison between the measured OH-LIF, CH$_2$O-LIF and experimentally-derived FRR (using k(T)*[OH]*[CH$_2$O], left hand plot) and the calculated concentrations and FRR at a=2500 s$^{-1}$ (right hand plot). The experimental and simulated FRR peak agree with each other, located at $f=0.11$ and $f=0.12$ respectively. The experimental LIF signals appear much broader compared to the simulated concentrations. A broader experimental signals compared to the calculated even the same burner configuration in experiment and simulation is used was reported by Mulla et al. [143] in freely propagating premixed laminar flame and Fayouq et al. [144] in counter premixed flames. The broadening in this work is thus expected to be larger, with the CH$_2$O FWHM 3 times broader compared to the opposed laminar flame calculation, which the same broader value was reported by Gordon et al. [71]. This broadening possibly comes from the collisional quenching and population distribution of the CH$_2$O that were not perfectly corrected due to insufficient data for this species. The corrections performed based on the semi-empirical temperature dependent as explained in Section 5.2.3.3 to the CH$_2$O signal however have improved the signal notably (see Figure 5-7). Furthermore, the comparison between the simulation and the experimental was not done with the same configuration, rather the experiment uses non-premixed free propagating jet flames, while the calculation was done in opposed diffusion flame. For the sake of clarification, a similar system for the OH-LIF was used for both work in this chapter and in Chapter 4, where comparison of OH signals was made
between the experimental and numerical simulation using the same burner configuration. The results agreed to each other then, and although the fuel and oxidiser mixtures are now different, the level of PAH contamination in the LIF signal is still believed to be minimal. Furthermore a high laser energy used in the CH$_2$O-LIF is likely to saturate the PAH fluorescence [108], in other words limiting the PAH signal. Additionally, the simulated peak heat release (see Figure 6-3 (right)) is located near the stoichiometric mixture fraction, $f_{st}$ while PAH interference is expected more at the fuel-rich region. Hence, even if the PAH were to exist, the CH$_2$O should not be falsely interpreted. The high strain rate used in this work also plays an important role in reducing the possibility of PAH formation; a high strain rate was reported by Manabu et al. [145] to highly reduce PAH by means of reduced residence time, which in turn reduces the probability of the extended chemical reaction. Thorough confirmation that the flames were clean from PAH interference, whilst important, was unfortunately beyond the scope of this work.

6.3.3 Instantaneous temperature, OH, CH$_2$O, gradient-squared temperature and reaction rate images

Figure 6-4 shows the instantaneous images of temperature, OH, CH$_2$O, temperature gradient and FRR for a 50 % O$_2$ flame at Re=18000 taken at x/D=5. All the images in the figure are simultaneous. The gradient-squared temperature, $|\nabla T|^2$ and FRR images are of interest to understand the correlation between the peak heat release (represented by forward reaction rate) and the thermal dissipation (represented by $|\nabla T|^2$) of the non-premixed oxy-fuel flame.

The edges of the reaction zone are overlapped onto every image to aid visualisation of the overlap regions. As expected, the reaction is predominantly found in the high temperature region and analysis shows the FRR peak correlates with temperatures above 1800 K. In the OH image, the outer layer of the FRR is shown to overlap near the OH peak. This outer layer is very sensitive in a way that if the CH$_2$O signal is broadened, it moves the outer layer of the FRR towards the oxidiser side and at the same time increases the thickness of the FRR. The inner layer of the FRR covers the entire OH signal at the fuel rich side as predicted by the simulation (see Figure 6-3).
The OH signal region that is not overlapped with the FRR at the far right (and far left) of the image is likely to be involved in the forward reaction of R287 which generates the secondary heat release peak discussed in section 6.3.1. Contrary to the OH in the oxidation zone, the FRR does overlap the outer edge of the CH$_2$O, and covers quite a large fraction of the CH$_2$O signal. In the $|\nabla'|^2$ image, the peak FRR, as expected lies in the low $|\nabla'|^2$ region and its edges (white solid lines) correspond well with the high $|\nabla'|^2$ region (shown in yellow). This is the opposite of premixed flames where FRR peak was reportedly found predominantly in the high $|\nabla'|^2$ region [142].

Figure 6-4: Instantaneous images of temperature, OH, CH$_2$O, gradient-squared temperature and reaction rate of 50 % O$_2$ flame at jet Re=18000 at x/D=5. The size of each image is 8 mm x 28 mm, and the nozzle centreline is marked with a vertical white dashed line. The region of forward reaction rate of [OH][CH$_2$O] is overlapped on each image, represented by solid white lines.
6.3.4 Scatter plots of FRR vs temperature and FRR vs gradient-squared temperature

Figure 6-5: Scatter plot of forward reaction rate of [OH][CH\textsubscript{2}O] vs temperature at (a) Re=12000 and (b) Re=18000 and forward reaction rate vs gradient-squared temperature, $|\nabla T|^2$ at Re=12000 (c) and Re=18000 (d). The scatter plots are from 50 % O\textsubscript{2} flame at 25 mm.

Figure 6-5 (a) and (b) show the scatter plots for FRR vs. T, and FRR vs. $|\nabla T|^2$ at Re=12000 (a) (c) and Re=18000 (b) (d). All points in Figure 6-5 represent samples taken from the right hand side reaction region (inside the solid white lines on the right of the image shown in Figure 6-4), at x/D= 5 using the same strategy presented in section 5.3.4. As such the samples are only taken from one pixel row along half of the image so the result here is comparable to those discussed in Chapter 5. Considering FRR vs T, the plots representing different Reynolds number are seen to give a similar behaviour. It is seen that some of the
samples at Re=18000 are higher compared to Re=15000. As discussed earlier in this thesis, a higher Reynolds number would enhance the fuel/oxidiser mixing by means of intense turbulence fluctuations. This in turn would increase the reaction rate between the species.

In the single-shot image shown in Figure 6-4, the peak FRR appeared at a lower $|\nabla T|^2$ region. For the FRR vs. $|\nabla T|^2$ scatter plots, this trend is consistently seen across the whole dataset of images. Increase in Reynolds number results in more scatter, with still more reaction (FRR) existing in the higher $|\nabla T|^2$ region, but the shift is not pronounced.

The difference in peak FRR values is further analysed by plotting the probability density function, excluding the pixel of zero values as shown in Figure 6-6. The 50 % O$_2$ and 55 % O$_2$ flames at different Reynolds number (Re=12000 and Re=18000) at $x/D = 5$ (25 mm) and $x/D = 10$ (50 mm) are plotted to identify the effect of increasing these parameters (O$_2$, Re and measurement locations). As explained in the previous paragraph, there is no difference observed in terms of the peak values of FRR in the 50 % O$_2$ flame for both Reynolds number at $x/D=5$. Figure 6-6 (a) also reveals the reaction rate is predominantly less intense; the peak of PDF is centred at around 4 (arbitrary units). In the downstream position at $x/D=10$, the PDFs have evolved slightly. Increasing Reynolds number has shown to shift the PDF profile peak and at the same time increasing the presence of high FRR regions. The same trend was also observed in the laminar flame calculation using CHEMKIN, where lower strain rate (i.e.
the Re=12000 case) has lower FRR/ HRR than higher strain rate. For the 55 % O2 flame, the change in Reynolds number is shown to start behaving differently even at 25 mm, by means of shifting the peak of the PDF to a higher FRR. This could be due to a higher concentration of O2 which increases the reaction rate beyond the 50 % O2 flame. At x/D=10, the difference becomes more obvious. For both flames, the plots show an increase of FRR for the measurement taken further downstream. This is because at downstream positions, turbulence will become relatively more important compared to diffusion, and a mixture-average diffusion model becomes more appropriate. Such a result was predicted using the laminar flame calculation; the mixture average diffusion model produced higher FRR compared to a calculation that takes into account the diffusivity of each species.

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<th>50 % O2 flame</th>
<th>55 % O2 flame</th>
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<tr>
<td></td>
<td>Re=12000</td>
<td>Re=18000</td>
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<tr>
<td>x/D=5</td>
<td>1.11%</td>
<td>1.31%</td>
</tr>
<tr>
<td>x/D=10</td>
<td>2.57%</td>
<td>2.61%</td>
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Table 6-1: Local extinction percentage (number of pixels with FRR=0 to the number of sampled pixels) in 50 % and 55 % O2 flames at different Reynolds number, at x/D=5 and x/D=10

A zoomed in view of the scatter plot near the x-axis in Figure 6-5 (b) shows some samples apparently in the reaction zone giving a zero value, (FRR=0) which is evidence of local extinction, potentially resulting from cold gases diffusing into the reaction region. Here, local extinction is defined as an absence of one (or both) of the fluorescence signals inside the reaction region. A careful post-processing with appropriate threshold was used for both fluorescence signals, and given that both signals are far broader compared to their simulated counterparts means that false positive extinction events are unlikely. The frequency of the local extinction events in all images has been assessed to compare the effect of increasing the O2 in the oxidiser, Reynolds number and the measurement location. For a fair comparison between these different cases, the total of zero pixels, FRR=0 (representing local extinction as illustrated in Figure 6-5 (b)) is divided by total number of pixels in the reaction region and presented as a percentage in Table 6-1. As observed in section 5.3.4, increasing the Reynolds number increases the probability that the flame is locally extinguished. Additional O2 in the oxidiser stream improves the combustion and reduces the localised extinction probability due to increased reaction rate. The downstream
region has been shown here to experience more frequent local extinction, which is almost double the upstream value.

This result (higher probability of local extinction in downstream position) determined using ‘zero pixel value of FRR’ is in agreement with the localised extinction trends determined using the OH ‘breaks’ visualisation in section 5.3.1. Note that good quantitative agreement could also be found between the results in this chapter and the polarisation/depolarisation method of chapter 5 by setting the threshold temperatures in the latter to an appropriate value (for 50 % O₂ oxidiser, threshold temperature = 1800 K). The laminarisation effect that was seen in all the flames at the upstream position further supports these findings, since laminarisation would tend to prevent any “break up” of the flame front whilst still allowing localised extinction to take place downstream. These three different methods: 1) zero pixels value of FRR, 2) OH ‘breaks’, 3) partially burned gases using temperature data, could be used to determine the local extinction, however the first two methods are seen more reliable without the need of additional assumption as compared to the third method where the assumption on the threshold temperature value need to be made.

6.4 Conclusion

Simultaneous measurement of temperature, OH- and CH₂O PLIF has been used to explore heat release in non-premixed oxy-fuel flames. Initially, an analysis using a laminar flame simulation was performed to understand heat release chemistry in a non-premixed flame. It was revealed that the high temperature flame typical of oxy-fuel combustion produces higher heat release in the secondary oxidation zone due to the forward reaction of R35 and R287. The laminar calculation revealed that the use of the forward reaction rate product k(T)[CH₂O][OH] underestimates the total heat release at lower strain rate, however it improves to give almost 80 % of the true heat release value at a higher strain rate of a=2500 s⁻¹ calculated using a multi-component diffusion model (which, importantly, is more consistent with the expected strain rate of the flame in this work).

Although the formaldehyde concentration cannot be fully quantified due to missing quenching partner data, care was taken to perform all corrections for temperature fluctuations as accurately as possible. For example, the k(T) parameter required for FRR was
characterised with some care. Despite this, a comparison of measured and simulated [OH], [CH₂O] and FRR showed that the experimental distributions were somewhat broader. It has not been conclusively proven, but this is probably not due to PAH interference and may instead be related to a difference in geometry; the simulation is performed under opposed flow where the experiment uses co-flowing fuel and oxidiser. Despite these provisos, the peak of the measured FRR nevertheless showed excellent spatial correlation to the calculated FRR peak, making FRR a suitable indicator for heat release rate (HRR) localisation whilst also being moderately useful for quantification.

Following data analysis, the HRR was shown to increase at the downstream region compared to the upstream. Increase of Reynolds number has also been shown to increase the HRR, with the downstream region showing an obvious difference while upstream is less pronounced. Further, the HRR signal was shown to mainly exist in the low temperature gradient region.

Localised extinction defined to be a zero value in HRR in the reaction region due to absence of one or both fluorescence signals was determined for all test cases. It was revealed that higher Reynolds number and measuring in downstream regions increases the probability of local extinction. The addition of O₂ however increases reaction rate, and lowers the probability of local extinction. These findings are largely in agreement with the analysis conducted in Chapter 5, but the consideration of heat release has allowed improved physical understanding; it appears that the threshold temperature values used in the Chapter 5 analysis were too conservative.
7 Spatial analysis of dissipative structure in oxy-fuel flames using high-resolution Rayleigh

7.1 Introduction

Dissipation is the smallest scale in turbulence. Understanding of flame structure has to go beyond knowledge of chemical reaction, species concentration or temperature. A perfect chemical reaction model but with little understanding of what happens on the smallest scale might predict a misleading numerical result. Hence, this chapter which investigates dissipative structure can be considered a complementary study to the earlier presented results in Chapter 5 and 6. Phenomena on the smallest scales of the turbulent non-premixed oxy-fuel flames are analysed in this chapter, using a high spatial resolution setup.

Molecular mixing and chemical evolution in non-premixed turbulence flames are governed by the scalar dissipation rate, \( \chi \) given by

\[
\chi = 2D(\nabla f, \nabla f)
\]  

Where \( D \) and \( f \) are the mass diffusivity and mixture fraction respectively. Though several studies of scalar dissipation measurement have been performed before, for example by Frank et al [51] and Fielding et al. [146] in two-dimensional and Karpetis et al. [147] in one dimension, the SNR is rather low especially when Raman scattering or LIF techniques are applied. A low spatial resolution in Raman scattering also limits the structure that could be resolved [148]. The determination of scalar dissipation also demands a joint scalar measurement approach to generate reliable data for the mixture fraction gradient.

Thermal dissipation structure which is characterised by the flame temperature could be an alternative way to compare experimental data to numerical simulation at the smallest scale. Thermal dissipation, \( \chi_T \) is given by

\[
\chi_T = 2\alpha(\nabla T', \nabla T')
\]
where $\alpha$ and $T'$ are the local thermal diffusivity and temperature fluctuation respectively. The single shot temperature fluctuation, $T'$ is given by

$$T' = T - T_{\text{mean}}$$

(7.3)

where $T$ is the single shot temperature while $T_{\text{mean}}$ is the average temperature. As the temperature data can be collected using Rayleigh scattering, high SNR and spatial resolution can be achieved. Thermal dissipation structure has been successfully inferred using Taylor’s hypothesis by Wang et al. [149] from time series temperature data in one-dimension. This method however is only applicable in the downstream region where turbulence dominates. As has been shown in previous chapters, by means of temperature and OH images, the flame structure at the near field region is strongly influenced by the laminarisation effect. The velocity and scalar fields in this region are also significantly affected by heat release [46]. Hence understanding gained from a non-reacting jet cannot be transferred to a reacting jet at the near field as it might in the far field (downstream) region.

Thermal diffusivity was scaled to temperature by Wang et al [150] and it was reported to be approximately represented by

$$\alpha \propto T^{1.72}$$

(7.4)

Though the temperature images inferred from the Rayleigh scattering signals have high SNR, image processing using equation (7.2) would generate unacceptably noisy images. The variation in the local thermal diffusivity is therefore often neglected to avoid introducing noise that would compromise the accuracy of thickness measurements. The omission of $\alpha$ does not have any significant impact on the layer thickness as the variation of the thermal diffusivity generally occurs at a larger length scales than considered in this chapter [20].

In this chapter, the thermal dissipation thickness at different axial positions with varying jet Reynolds number and co-flow oxidiser oxygen concentration are analysed. The analysis in this chapter is believed to extend the knowledge of the layer thickness in oxygen enhanced oxy-fuel flame. The dissipation analysis was previously performed by Kaiser and Frank [20,
however, the comparison was only done at different axial positions in ‘Flame DLR-A’\textsuperscript{10}. The thermal diffusivity is neglected to ensure noise is maintained at the minimum level, which simplifies equation (7.2), with the remaining term the squared temperature fluctuation, $|\nabla T'|^2$

$$|\nabla T'|^2 = \left( \frac{\partial T'}{\partial r} \right)^2 + \left( \frac{\partial T'}{\partial x} \right)^2$$

(7.5)

Where $\left( \frac{\partial T'}{\partial r} \right)^2$ and $\left( \frac{\partial T'}{\partial x} \right)^2$ are the radial and axial contributions, respectively. Note that equation (7.5) uses temperature fluctuation field, $T'$ rather than temperature field, $T$ that was used in Chapter 6, for the squared-gradient temperature analysis. This is because $T'$ captures the smallest turbulent structures, and was reported to account nearly all dissipation involves in the flame [20].

### 7.2 Methodology

#### 7.2.1 Experimental setup

In this experiment, temperature was measured using polarisation/ depolarisation Rayleigh. This has been extensively discussed in Chapter 5. The same Rayleigh technique is used here, however a high resolution measurement requires some changes in the Rayleigh light collection setup and these will be discussed now.

The polarised signal was collected using un-intensified interline transfer CCD camera (LaVision Imager Intense), coupled with an 85 mm f/1.2 Mitakon Speedmaster lens. For the depolarised signal, it was collected using an ICCD camera (LaVision Imager Intense plus IRO using GaAsP photocathode). Another Mitakon Speedmaster lens of the same specifications was used to collect the depolarised signal, and it was coupled with a 2x tele-macro adapter. This serves to compensate the inherent magnification of the IRO to ensure a matching magnification on both cameras. The geometric resolution of the system was 21.6 $\mu$m x 21.6 $\mu$m for every pixel, with a line spread function (LSF) measuring 25 $\mu$m full width half maximum (FWHM). The LSF takes into account the optics and array detector and was

---

\textsuperscript{10} DLR-A flame has a fuel mixture of 22.1\% CH4, 33.2\%H2 and 44.7\%N2. The jet velocity was set to 42.2 m/s, with Re=15200.
characterised using a knife-edge technique following the method presented by Clemens [152]. The FWHM of the laser thickness is around 95 μm. Though the laser thickness is coarser than the in-plane resolution (three times greater in this system) this has been reported to have only minor impact on the in-plane gradient measurement [20] and thus the influence of the laser sheet thickness is neglected.

The fuel/oxidiser mixtures used here are the same as described in Chapter 5. In this experiment, the Reynolds number and oxidiser oxygen concentration were varied and flame images were collected at x/D = 5 and x/D = 10. The test cases are summarised in Table 7-1.

<table>
<thead>
<tr>
<th>Flame</th>
<th>x/D</th>
<th>O₂ %</th>
<th>Reynolds number</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>25 mm</td>
<td></td>
<td>Re=12000</td>
</tr>
<tr>
<td>A2</td>
<td>50 mm</td>
<td>50 %</td>
<td></td>
</tr>
<tr>
<td>A3</td>
<td>25 mm</td>
<td></td>
<td>Re=18000</td>
</tr>
<tr>
<td>A4</td>
<td>50 mm</td>
<td></td>
<td></td>
</tr>
<tr>
<td>B1</td>
<td>25 mm</td>
<td>55 %</td>
<td>Re=12000</td>
</tr>
<tr>
<td>B2</td>
<td>50 mm</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 7-1: Test cases for the high-resolution Rayleigh temperature measurements

### 7.2.2 Data processing

Thermal dissipation thicknesses are evaluated from 500 single-shot temperature images for every test case. However, a single-shot temperature image is likely to be affected by noise especially in high temperature regions. The signal to noise ratio (SNR) for this system is considered high, about 62 in oxidiser at 293 K but it reduces at an elevated temperature. Noise suppression is important to get an accurate result. The noise was reduced by adopting inhomogeneous smoothing with an isotropic Gaussian kernel, similar to Frank and Kaiser [20] as explained in Section 6.2.2, equation (6.2).

In the flames investigated here, the Gaussian kernel size, σ_T, could vary between about 1 to 6 pixels, translating to about 22 μm to 130 μm. The reference kernel size, σ₀, was determined to be 1.2 pixels after an analysis was performed using a homogenous filter in a
low temperature region. A larger value would generate too much attenuation of important structure.

Figure 7-1 illustrates the step-by-step process to measure the dissipation layer thickness. All the images are first filtered using the smoothing technique discussed earlier; Figure 7-1 (a) shows an example single-shot image after adaptive smoothing. 500 images are then averaged, followed by a determination of the temperature fluctuation, as shown in Figure 7-1 (b) and (c) respectively. The axial and radial squared gradient fluctuation contributions are then calculated, and summed to give the total squared gradient fluctuation, $|\nabla T'|^2$, which consists of a number of closely packed layers as shown in Figure 7-1 (d).

Each layer centreline is then determined. However, to reduce the uncertainty in locating the local maxima of the layer, a “special $|\nabla T'|^2$” image was used where the same temperature
image (Figure 7-1 (a)) is smoothed with twice the kernel size calculated earlier (It should be noted that this special image is only used to locate the centrelines and not used for further processing). Figure 7-1 (e) shows an example of the determined centrelines (red lines) overlaid on the $|\nabla T'|^2$ image of Figure 7-1 (d). The thickness of each layer is then determined at every point along the centreline in the normal direction as shown in the zoomed image of Figure 7-1 (f); each normal is marked with a yellow line. Following the approach applied by Frank and Kaiser [20] and the references therein, the thickness is measured as a full width at 20 % of the local maximum. Therefore, at every point along the centreline, the distance from the local maximum in the positive and negative directions of half width at 20 % local maximum are identified separately, and each distance is multiplied by two to form the full width. To reduce uncertainty in the thickness measurement, only profiles which decay monotonically to 20 % of the maximum are measured and included in the analysis of dissipation thickness that follows.

**7.3 Result and discussion**

**7.3.1 Mean contribution of $|\nabla T'|^2$ and $|\nabla T|^2$**

All the results presented in this section are compiled from 500 images for every condition, from 250 x 619 pixels representing a 5.4 mm x 13.4 mm region in the flame.

Figure 7-2 shows the mean values of squared temperature (black lines) and temperature fluctuation (red lines) gradients contributed from the axial, radial and (axial + radial) dissipations from two different jet Reynolds number, at Re=12000 and Re=18000 at x/D = 5 and x/D = 10 for 50 % O$_2$ flames. At x/D = 5, both of the flames at Re = 12000 and Re = 18000 show similar profiles for the temperature and temperature fluctuations gradients. Re = 12000 is seen to produce slightly lower magnitude of gradient compared to the higher Reynolds number case. This is expected because as the Reynolds number is increased, the turbulence fluctuation also increases and there is more entrainment of cold gases into the reaction zone, generating higher fluctuations in the temperature and higher gradient magnitudes. The gradient profiles for Re = 12000 are also seen to shift towards the co-flow due to a broader reaction zone as discussed in Chapter 5.
In both test cases, the radial gradients \( \left( \frac{\partial T}{\partial r} \right)^2 \) and \( \left( \frac{\partial^2 T}{\partial r^2} \right) \) contribute more to the total dissipation compared to the axial. For \( |\nabla T|^2 \) in the outer reaction region i.e. between the reaction zone and the co-flow at around \( r/m \approx 0.006 \), \( \left( \frac{\partial T}{\partial r} \right)^2 \) contributed almost entirely to the total value of the dissipation. This is the region near which the highest temperature is located in the flame. Here, the dissipation structures are mostly positioned vertically.

The \( \left( \frac{\partial T}{\partial r} \right)^2 \) and \( \left( \frac{\partial T}{\partial x} \right)^2 \) profiles enable the degree of laminarisation effect to be analysed. A high ratio of \( \left( \frac{\partial T}{\partial r} \right)^2 \) to \( \left( \frac{\partial T}{\partial x} \right)^2 \) indicates the laminarisation effect is dominant, while a small ratio indicates turbulence becomes appreciable. Figure 7-2 shows the laminarisation effect dominates at the downstream region, and reduces at the downstream position. The laminarisation acts to reduce the entrainment of the cold co-flow into the reaction region. As the flame is somewhat laminar in this region, it should also
produce a relatively low fluctuation, $|\nabla T'|^2$. Between nozzle centre to $r/m = 0.004$, \[ \left( \frac{\partial T}{\partial x} \right)^2 \] and \[ \left( \frac{\partial T'}{\partial x} \right)^2 \] are seen to increase. Kothnur et al. [153] has also reported this in their study and suggested the reason is due to the 45° orientation of the compressive strain that is induced by the heat release from the flame.

Further downstream at $x/D = 10$, the magnitude of the gradients reduces to less than half of the $x/D = 5$ values. The radial and axial gradients are getting closer in strength, suggesting that turbulence has started to play its role. The peak ratio of \[ \left( \frac{\partial T}{\partial x} \right)^2 \] and $|\nabla T|^2$ at the fuel rich to the fuel lean region for $Re=12000$ is also seen to reduce as compared to the upstream region. These peaks however are still noticeable for $Re = 18000$.

In order to capture the turbulence structure and the heat dissipation structure in the flame, the analysis should be based on the temperature fluctuation gradient rather than the temperature gradient. However, the mean gradient of temperature $|\nabla T|^2$ also contributes to the total dissipation. This is true at the $x/D = 5$ at the peak of temperature fluctuation. The mean gradients however are shown to dramatically reduced in the upstream position, at $x/D = 10$. 
7.3.2 Thermal dissipation structure

Figure 7-3: PDF of Layer thicknesses conditioned on temperature range of 300 K - 1700 K. Each PDF line represents a temperature interval of 200 K.

Figure 7-3 shows PDFs of layer structure width taken at x/D = 5 and 10 at Re = 12000 (top row) and 18000 (middle row) for the 50% O₂ flames and at Re = 12000 (bottom row) for the 55% O₂ flames. Each PDF represents the compilation of 500 images, and the layer thickness
is conditioned on the temperature, across a range of 300 K to 1700 K. Each PDF line represents a temperature interval of 200 K (e.g. the 600 K line includes data from profiles collected between 500 K and 700 K). It should be noted that there is a spurious peak at a layer thickness, $\lambda_D$ of 100 $\mu$m due to the detection of a small amount of beam non-uniformity by the algorithm. The non-uniformity is virtually undetectable in the images and should be ignored for the purposes of this analysis.

From the PDF results, the peak in layer thickness (labelled as $\lambda_D$ at $T = 600, 800, 1200 K$ in the figure) shifts to the right as temperature increases, suggesting dissipative layer thickness increases proportionally with temperature. Comparing two different axial positions (left column to the right), the PDFs also suggest that the layer thickness tends to increase downstream (comparing the PDF at the same temperature interval), although thickening due to the increase of temperature is more significant (i.e. comparing two PDFs at the same axial position). The PDF profile at higher temperature is also wider than those at lower
temperature, indicating broader range of different thickness of the thermal layer at high
temperature for example the FWHM is 112 μm and 196 μm at 400 K and 1600 K respectively
for the top-left PDF in Figure 7-3. As expected, increase in Reynolds number will result in
decreasing the dissipative layer thickness. Referring to the top and bottom rows of Figure
7-3, increasing O₂ concentration in the oxidiser leads to a tiny impact on the layer thickness.
However, it should be noted that the difference in the O₂ concentration is only 5 % leading
to an adiabatic temperature difference of 160 K. Taking this conditional sampling approach
into account, any changes in the thickness are hardly noticed.

For the layer thickness measurement result to be usable – for example to model it
numerically, the layer thickness dependency on temperature should be expressed
analytically. Figure 7-4 shows the result of applying a power law to the layer thickness PDFs
of Figure 7-3 where the scaled thickness, \( \lambda_o^* = \lambda_o (T/T_0)^{-n} \) following the scaling law
applied by Frank and Kaiser [20]. \( T_0 = 400 \) K and \( n = 0.3 \) have been used which result in a
good overlap (mainly at the layer thickness PDF peaks) for all the test cases. It should be
noted that the wings on the left side of the scaled thickness (which were at \( \lambda_o = 100 \) μm) are
the results of the previously mentioned beam non-uniformity. This artefact becomes more
pronounced at high temperature in downstream regions because high temperature leads to
a lower SNR, and at downstream locations the high temperature region is broader giving
more opportunity to generate artificial structures. Analysis of the main peak of the PDF
remains unaffected, however.

7.4 Uncertainty analysis

Knowledge of the spatial resolution of the system is important to ensure the derived layer
thickness from the temperature images can be accurately quantified. Wang and Clemens
[148] conducted a study to analyse the relative error of a derived scalar from acquired
images for a given spatial resolution. They suggested that in order to ensure < 10 % error in
scalar structure thickness (measured at full width 20 % maximum), the measurement’s
spatial resolution should be 6.1 times smaller than the structure. In this work, the FWHM of
the optical system resolution was identified using a knife-edge technique, and the
smoothing applied to the images was also considered in determining the error. As the kernel
size of the adaptive smoothing increases with temperature, the relative error in a layer structure that has the same thickness, but different temperature will be change. As an example, referring to Figure 7- 3, to keep the error below 10 %, the layer thickness should be above 97 μm and 184 μm for the PDF lines of T=400 K and 1200 K respectively. It is therefore estimated that the peak layer thicknesses discussed in this chapter are accurate to better than 10 % error.

7.5 Conclusion

A detailed analysis of the dissipation structure of an oxy-fuel co-flow diffusion flame has been performed with varying jet Reynolds number and oxidiser oxygen concentration at axial positions of x/D = 5 and x/D = 10. Evidence of the laminarisation effect in the near-field region was found, judged by the relative contribution of radial and axial gradients and fluctuations (radial components being markedly stronger). PDFs of layer thickness conditioned on temperature were generated at 200 K intervals over a temperature range of 300 K – 1700 K. The PDFs show that the dissipative layer thickness is directly proportional to the temperature. The results also suggest that layer thickness increases with height/ axial position. Increase of the jet Reynolds number has been shown to decrease the layer thickness, while increasing the oxygen concentration by 5 % in the oxidiser led to a minor increment to the layer thickness, perhaps because the adiabatic temperature difference is only 160 K. A simple power law was used to scale the layer thickness PDFs. The scaled thickness is calculated using $\lambda_0' = \lambda_0 \left(\frac{T}{T_0}\right)^{-n}$ and values of $T_0 = 400$ K and $n = 0.3$ were shown to work well, enabling this result to be integrated in future modelling efforts.

This chapter has resolved the fine scale of the dissipation structure of oxygen enhanced oxy-fuel flame. This chapter completes this journey into the detail of the structure of oxygen enhanced oxy-fuel flames and extend the work performed by Kaiser and Frank [20, 151] into a different combustion mode.

The main difference that could be drawn from this investigation compared to the work performed by Kaiser and Frank are as follow: 1) for a particular test case presented in Figure 7- 3, the percentage difference between the peak layer thickness at temperature of 600 K and 1200 K is about 25 %, while in their work, the percentage difference is about 45 %.
comparing the peak at 600 K between two axial positions at x/D = 5 and x/D = 10, the thickness was reported 36 % thicker at downstream compared to the upstream position, while in this work, the difference is only about 7 %. 3) The constant, \( n \) for scaled layer thickness in this investigation was found to be 0.3, while in their case was 0.75. These differences possibly come from the different geometry and fuel/ oxidiser mixture used. Conclusive reasons to support these however require at least the burner geometry to be the same. Further, In the case of increasing O\(_2\) concentration, the thickness was hardly changed. It would be interesting to see if further increase of O\(_2\) would make any change and this could be a subject of future study.
Conclusions and future prospects

8.1 Conclusions

This thesis detailed a study of the structure of oxygen enhanced oxy-fuel flames using a spread of complementary laser diagnostics. The investigation included non-premixed laminar, normal diffusion and inverted diffusion flames established on a freely propagating co-flow jet burner, and employed two-dimensional laser-based imaging techniques.

Since oxygen-enriched flames burn at a higher temperature, they require a slightly different treatment compared to their air-fed counterparts. To give an example, thermal decomposition leads to significant production of several intermediate species that are often neglected in “normal” combustion studies. The addition of CO$_2$ as a diluent in oxy-fuel flames complicate matters further as this species has different properties compared to N$_2$.

Therefore, the initial phase of this work was a study which investigated the chemical reaction and the impact of those reactions and the generated species on the best choice from the available laser diagnostic techniques options to accurately measure combustion-relevant quantities, mainly temperature and intermediate species concentrations.

The Rayleigh scattering technique was ultimately chosen. Although Raman scattering can detect typical major species, it stumbles when faced with intermediate species such as O, H and OH. These species account for more than 5\% by weight and nearly 4.5\% of the Rayleigh signal in the oxy-fuel flames discussed in this thesis. Furthermore, other heavy species such as C$_2$H$_4$, C$_2$H$_6$ and C$_3$H$_8$ are often neglected in analysis of Raman data. Though these “heavy” species are only 0.8\% of the overall concentration, they contribute almost 3\% to the overall Rayleigh signal. In air-like temperature oxy-fuel flames, a joint Rayleigh/Raman scattering technique might be favourable as the aforementioned species are present at the minimum level and can be safely neglected. At high temperature, however, using Raman species concentration to correct the Rayleigh signal would cause an error of 7.5\%, with extra error anticipated when the (in)accuracy of each Raman species’ concentration accuracy is taken into account. The Rayleigh technique is “blind” to species but the composition of the mixture must still be known to derive an accurate temperature.
A decision was made to tackle the compositional uncertainty in two different ways, as illustrated in chapters 4 and 5, whilst benefitting from the increased signal levels (and thus improved SNR) of Rayleigh. The high SNR also enable two-dimensional Rayleigh signal to be collected for a better flame structure interpretation.

Three main experimental campaigns were conducted in this work and the results have been presented in four chapters. The main findings of these chapters are presented next:

**Chapter 4**

In the first part of the chapter, an investigation was made to understand the feasibility of comparing the experimental signal collected employing Rayleigh scattering technique to the numerical calculated Rayleigh signal. The comparisons were made in term of Rayleigh ratio, which this Rayleigh ratio is easily be calculated from the species and temperature information calculated in the numerical simulation. The comparisons were made in the laminar and turbulent non-premixed flames at two and three downstream positions respectively. Laminar flames were first compared where the sensitivity studies were performed to find the most suitable radiation and diffusion models that can represents the oxy-fuel flames based on the recorded signal. The Soret effect was also investigated. From the analysis, it was shown that the radiation model influence the temperature more than the species distribution. However, for the Soret effect, the temperature does not vary much with or without Soret effect considered. But, the species distribution was shown to change significantly. The Rayleigh ratio, which highly affected by the temperature hence does not change much due to small change in the temperature. This shows that the comparison made using Rayleigh ratio is more sensitive than comparison made using temperature profile alone particularly when considering the Soret effect sensitivity. This information gained from laminar flame comparisons would be very useful when further analysis on the turbulent flames are required.

The comparison in the turbulent flames were then performed. The structure of the two-dimensional were able to be compared side by side and further mean and rms comparisons were made. Though the numerical model incorporated all the information gained in the laminar flames as explained earlier, the numerical model seems to underestimate the Rayleigh ratio in the upstream region, however improves at the most downstream region.
This is expected as in the most upstream region, complex flames behaviours were reported by many studies. Further, the signal fluctuations at several locations in the image domain were made to test the feasibility of the comparison using Rayleigh signals.

In the second part, another signals comparison and validation between the experiment and numerical are made. This investigation involved a joint Rayleigh and OH-PLIF measurement. Again, in this experiment, with the knowledge of temperature and species distribution, the numerical calculated OH concentration data was used to predict the experiment OH signal. Comparison of the OH and Rayleigh signals were made in normal and inverted oxy-fuel flames configurations, where the jet flow was kept at the same Reynold number in these two configurations. The inverted flame Rayleigh ratio was seen to have a narrower structure compared to the normal flame in the mean profile, with almost similar Rayleigh ratio peak indicating similar temperature reading. The OH for IDF flame also shown to be narrower than the NDF, but the peak difference was not able to evaluate as the signal is not quantitative. The comparisons were made at a height of 67 mm where the turbulent has dominated the flow to avoid boundary condition effect. Further, joint PDF of the OH and Rayleigh ratio were made to further verify the model and applicability of this signal comparison technique.

To summarise, this investigation to directly validate the numerical simulation using experimental measured signal by employing Rayleigh scattering has been shown feasible in laminar and non-premixed oxy-fuel flames. Rayleigh ratio signal has been shown to be more sensitive compared to the Rayleigh signal derived quantity (temperature), for example in Soret effect sensitivity analysis. Furthermore, the ‘traditional’ Rayleigh requires the Rayleigh cross section to be constant, otherwise relatively high error should be expected. In the oxygen enhance oxy-fuel flames, the constant Rayleigh cross-section is impossible due to the thermal decomposition at high temperature. In air-like oxy-fuel flame temperature, joint Rayleigh/ Raman measurement could be performed. The existence of high intermediate species concentration in high temperature however make the Rayleigh/ Raman technique prone to inaccuracy as the intermediate species could not be detected by Raman. Furthermore, one-dimensional imaging using Rayleigh/ Raman technique would make the validation across entire simulation field of view impossible. This technique also offers a wide range of fuel-oxidiser mixtures and at the same time keeping the uncertainty
at the minimum level. The experimental complexity is also reduced without the need of joint measurement to correct the temperature-derived quantity. The joint measurement of two quantities (temperature and OH) were also investigated to verify the technique and at the same time prove the applicability and feasibility of the technique to be applied for numerical verification purpose.

Chapter 5

In this chapter, another Rayleigh scattering technique called polarised/ depolarised Rayleigh was explored. Using information from polarised/ depolarised signals, the effective Rayleigh cross section can be predicted based on the numerical calculated data, and hence the correction can be made to the polarised Rayleigh signal to infer the flame temperature. The temperature measurement was performed simultaneously with hydroxyl (OH) and formaldehyde (CH$_2$O) PLIF in normal diffusion flames. The objective was to investigate the effect of varying O$_2$ in the oxidiser and varying the jet Reynolds number to the flame structure.

First comparison was made towards the flame temperature structures using the single shot images at different axial locations. At the upstream region, lower Reynolds number was seen to be influenced by the laminarisation effect. However, this effect decreased with the increasing of Reynolds number where wrinkled structure was seen as a result of turbulent domination. The mean and rms results show that varying the O$_2$ from 50 % to 55 % in the oxidiser concentration do not make any significant effect to the flame structure other than different magnitude of temperature, OH and CH$_2$O signals.

The OH signals were further analysed. The post-flame zone thickness indicated by OH was measured to determine the effect of increasing O$_2$ and Reynolds number. The result shows that increasing of Reynolds number results in thinner OH while the increasing of O$_2$ in the oxidiser was shown to broadened the post-flame zone. The PDF of OH thickness was then compared at different axial locations. At the upstream region, a narrow PDF was shown due to the laminarisation effect and get broadened with the increase of axial position as the flame has started to get influenced by the turbulence.
Scatter plot of temperatures for Re=12000 and Re=18000 at x/D=5 and x/D=10 were analysed and compared to the calculated temperature at different strain rates and diffusion models. At the upstream region, the calculated temperature was shown to underestimate the measured temperature especially at lower Reynolds number. This however improved when the Reynolds number or the axial position is increased. From the scatter plot, the local extinction was seen to happen more frequent at higher Reynolds number, at upstream region (based on the outlier points at relatively low temperature). Localised extinction was further assessed using narrow band of mixture fraction centred at highest local temperature. Temperature threshold was first set to be 1500 K for 50 % O_2 flames. All flames show a very low local extinction probability with the Re=18000 at x/D =5 being the highest. However, if the threshold increased to 1800 K, the probability is highest at Re=18000 at axial location of x/D=10 which valued at about 4 % while at x/D=5 for the same Re valued at 2.3 %. Hence, this shows that this method is very sensitive and highly depends on the threshold temperature. The analysis made visually looking at the ‘break’ of OH signals also shows the same trend as the latter result.

As has been discussed in detail in this thesis, oxy-fuel flames behave differently compared to “normal” air-like combustion. Thermal dissociation and production of intermediate species limit the applicability of common laser diagnostics techniques, especially in measuring the temperature. Two possible approaches based on Rayleigh scattering have been described, critically discussed, tested and presented. Choosing an appropriate technique should be based on several factors and the merit/ demerit of each approach:

1) Measurement error – From the measurement error perspective, errors from the experimental setup include beam non-uniformity, laser fluctuation (if the energy is not corrected) and also the Rayleigh cross section temperature sensitivity. These have been assessed to sum to around 4.7 %. For the temperature inferred from the polarised/ depolarised Rayleigh signals, the experimental error mentioned has to be included as it uses the same principle, however there is also a need to include the ‘analysis error’. Analysis errors are the error introduced if one derived the physical quantity from a measured signal. (Derived temperature requires additional information and assumptions about the flame.) From the analysis performed in this
thesis, the error might come from the strain rate and diffusion model used. The combine errors (experimental + analysis) from these sources summed to around 7%

2) Measurement purpose – For numerical validation purposes, direct signal comparison is highly effective. With the knowledge of temperature and species from the numerical calculation, the Rayleigh signal is relatively easily predicted. This saves experimental effort. However, if one wants to measure the temperature in the flame and use this to better understand the combustion phenomenon, the polarisation/depolarisation Rayleigh measurements can come close to the same accuracy.

3) Fuel/ oxidiser mixtures – In the study detailed in chapter 4, the Rayleigh cross-section variation for the fuel/ oxidiser mixture used in the Rayleigh ratio measurement was kept to moderate levels (±7%), but this was not necessary and a wider range of mixtures could be employed. By contrast, in the polarised/ depolarised technique, one has to make sure the normalised signal difference (see Figure 5-2) is not multivalued. To achieve a good signal difference, a significantly larger or significantly smaller Rayleigh-cross section molecule needs to be employed either in the fuel or oxidiser stream.

Chapter 6

In this chapter, further analysis of the oxy-fuel flames was performed to investigate the feasibility of using the product of [OH][CH\textsubscript{2}O] to represent heat release in oxy-fuel non-premixed flames. Heat release mainly relates to the reaction zone. Combustion instability, combustion noise and local extinction are all closely related to heat release information. Heat release localisation in a flame is therefore a popular subject in combustion. At a fundamental level, combustion is all about the conversion of chemical potential into thermal energy, further motivating study of heat release.

Analysis using the numerical calculated data revealed the heat release profile in non-premixed oxy-fuel flames consist of two peaks, one is called main oxidation and the other is called secondary oxidation zone. Most of the heat release contributes to the secondary oxidation zone were the forward reaction of \( H + O_2 + H_2O \leftrightarrow HO_2 + H_2O \) and \( OH + HO_2 \leftrightarrow O_2 + H_2O \). The product of forward reaction rate, FRR of [OH][CH\textsubscript{2}O] was shown to
underestimate the heat release at lower strain rate, however improves and covering 80 % of the heat release area at higher strain rate due to reduction of these reaction. From the numerical data, the HRR and FRR show good spatial correlation in the main oxidation zone.

The correlations between FRR and temperature as well as FRR and temperature gradient were analysed. The FRR was shown to start taking place at temperature of around 1250 K in the fuel rich region, while a steeper correlation was shown in the fuel lean region. Comparing the magnitude of the FRR at different Reynolds number, The FRR magnitude was shown higher in the higher Re compared to the lower one. From the FRR vs temperature gradient scatter plot, it was revealed that in this non-premixed flames, the FRR mostly happens at the low temperature gradient.

The local extinction analysis was also performed using the reaction rate signal. This was done to compare the results obtained in the Chapter 5. The result revealed the local extinction probability increases with the increase of axial position and Reynolds number. At an axial location of x/D=10, the Re=18000, 50 % O₂ flame has the probability of local extinction of 2.6 % and 1.31 % at x/D=5. The result also revealed an additional of O₂ concentration in the oxidiser improves the reaction rate, hence reducing the probability of local extinction.

Localised extinction analysis was performed using three different approaches in this thesis. To recap, the first approach was to visualise the OH images where the analysis was based on the OH ‘break’. This was done on ‘obvious’ local extinction. From the analysis of all images, the oxidiser with 50 % O₂, at 75 mm and Re=18000 was recorded to have the highest occurrence at 4 %, while at 50 mm axial location with the same Reynolds number and O₂ concentration this dropped to 2 %. Second, analysis was performed based on temperature data conditioned by a narrow band of mixture fraction located at the maximum average global temperature. In that analysis, if the threshold was set to 1500 K, the 50 % O₂ oxidiser at 25 mm with Re=18000 has the highest levels of extinction, at 0.7 %. However, if the threshold value is increased (still at 50 % O₂ in the oxidiser) to 1800K, the highest levels of extinction are found at 50 mm with Re=18000 using the 50 % O₂ oxidiser, with a frequency of 4 %. The same Reynolds number and O₂ concentration at the upstream position gives just 2.3%. Finally, localised extinction was determined using the reaction rate images with the
results specified in the previous paragraph. With the OH visualisation and reaction rate image methods, it is fair to say that these two approaches are more reliable for three reasons. First, from the OH image, the analysis was performed based on an ‘obvious’ break. False positives are essentially impossible. Second, in the reaction rate images, a combination of two quantities (OH and CH₂O) are used to generate one reaction rate image. It has been shown in the FRR vs mixture fraction plot (see Figure 6-3), that the OH and CH₂O is broader than the simulated quantities, likely forming broader reaction rate images. Hence, underestimating the reaction rate signal and generating a false positive (for extinction) is again unlikely. Finally, based on the temperature gradient analysis in the Chapter 7 (though the data was collected in a different run, the fuel/ oxidiser mixtures and the experiment environment are the same), it was shown that the radial component \((\partial T/\partial r)^2\) contributes almost entirely to the total temperature gradient in the upstream region, while the axial one \((\partial T/\partial x)^2\) is small. However, in the downstream region, the contribution is comparable from both. These different behaviours clearly show that there are more penetration/entrainment events of the cold oxidiser or fuel into the reaction zones downstream, which would be expected to increase localised extinction.

Chapter 7

Chapter 7 was devoted to the detailed analysis of thermal diffusion structures using high-resolution polarised/ depolarised Rayleigh scattering. This structure is critical to successfully model a flame as this dissipation region is where most of the reaction occurs. This analysis will definitely improve understanding of turbulent interaction in the oxy-fuel flames. The fuel and oxidiser mixtures were the same as in Chapter 5 to 6. Again, the analysis was performed to understand the effect of varying Reynolds number and O₂ concentration at different axial position, \(x/D=5\) and \(x/D=10\).

Generally, the result revealed that the thermal thickness is proportional to the temperature. The same trend was also shown when the O₂ concentration is increased, however the increment is small as the adiabatic temperature difference between the 50 % O₂ and 55 % O₂ flames are just 160 K. At higher axial position, the thermal layer was also revealed to be
thicker compared to those at upstream position, while the increase of Reynolds number reduces the thickness of the layer.

Finally, the PDFs layer thickness were scaled using a simple power law, \( \lambda'_d = \lambda_d \left( \frac{T}{T_0} \right)^{-n} \) and overlap nicely with \( T_0 = 400 \text{ K} \) and \( n = 0.3 \).

For comparisons with previously reported findings by Frank and Kaiser [20], flames considered in this thesis exhibit smaller thermal thickness. The percentage difference of peak layer thickness at 1200 K and 600 K was reported about 45 %, while in the oxy-fuel flames here are just around 25 %. The layer thickness at upstream position in their flame was reported to be 36 % smaller than the downstream (compared at the same temperature of 600 K). In this oxy-fuel flame however, the thickness percentage difference is significantly small, at about 7 %. Complex chemical process such as the laminarisation and diffusion influence at the near field regions hence need a lot more investigations for a better understanding.

### 8.2 Novelty

Chapter 4 presented an unusual way to validate a numerical model. The approach was based on the previously-published research by Connelly et al. [116]. The approach of validating temperature measurement by employing the Rayleigh scattering technique using Rayleigh ratio in oxy-fuel flame, in non-premixed laminar, turbulent normal diffusion flame (NDF) and inverted diffusion flame (IDF) to the best of author’s knowledge is a first. Furthermore, joint measurement for comparison/ validation of the Rayleigh ratio and OH-PLIF also improved and extended knowledge compared to previously-published research.

In Chapter 5, temperature measurement was performed using polarised/ depolarised Rayleigh. This technique were first presented by Fielding et al. [50] and Frank et al. [74] where the technique were presented under air-like flame temperature conditions. In this work, the technique was employed and extended to applications in oxygen enriched combustion. Fuel/ oxidiser mixture selection was tailored to make temperature measurement in non-premixed oxygen enriched oxy-fuel flames possible. The temperature combined with known mixture fraction measured using the technique was shown to enable
the temperature/composition (quenching) dependence of the OH and CH$_2$O-PLIF signals to be corrected. To the best of the author’s knowledge, this is the first time a two-dimensional temperature, and joint measurement of temperature, OH and CH$_2$O has been performed in an oxygen enriched oxy-fuel flame.

In Chapter 6, heat release in a non-premixed oxy-fuel flame was presented and this is also thought to be the first time such analysis using the product of [OH]x[CH$_2$O] has been performed in a non-premixed oxygen enhanced oxy-fuel flame.

In Chapter 7, thermal dissipation thickness analysis was performed based on the technique presented by Frank and Kaiser [20]. In the published research, a comparison of dissipation thickness at different axial locations were performed in a two-dimensional thermal analysis. In this work, a similar approach was used, but the application is in the oxygen enhanced oxy-fuel flames, and both Reynolds number and oxidiser O$_2$ concentration were varied.

**8.3 Future prospects**

Continuous advances in computing have enabled numerical models to simulate more complex flame structures that have not been achieved before. This advancement has also demanded that the experimentalist keeps up; to improve the capability of the measuring techniques. These improvements include the development of new techniques that enable more species to be measured, or even the improvement of available techniques to achieve higher measurement accuracy.

In the polarised/depolarised Rayleigh scattering measurements, the scattering cross section was determined by relying on the gas composition predicted by CHEMKIN as a function of mixture fraction. It is clear that in order to achieve the best measurement accuracy, that the correct strain rate and diffusion model should be employed. In the future, a complementary Raman measurement could be performed at different axial positions and/or Reynolds number to recover some species data, for example H$_2$O and CH$_4$. These two species might in fact be ideal candidates as they (combined signals of H$_2$O and CH$_4$) cover the whole flame profile (see Figure 5.1 (a) and (b)) from $f = 0$ to $f = 1$. Species profiles calculated using different strain rates and diffusion models could then be compared to the measured data, and the one that gives the best match used to analyse the Rayleigh signal. With this
additional Raman measurement, mixture fraction assessment in the fuel rich region could be improved compared to the current experiment, which had an estimated error of 5% (see Section 5.3.1 and Figure 5.2 (b)).

Further analysis of the re-ignition process in oxy-fuel flames using higher temporal resolution would be beneficial. Previously, investigation of the re-ignition process employed simultaneous measurement of temperature, OH and CH$_2$O PLIF and was only possible with a specific fuel due to the limitations of ‘traditional’ Rayleigh. Collecting temperature measurements using the polarised/ depolarised Rayleigh technique instead relaxes this limitation and enables a broader range of fuel/ oxidiser mixtures to be investigated, extending our understanding of combustion. Typical high-repetition rate laser systems may produce pulses sufficiently quickly (at kHz rates), but with no more than approximately 5 mJ per pulse, good SNR with Rayleigh scattering is impossible. However, the recent development of HEPBLS (high energy pulse burst laser systems), as mentioned in chapter 2, makes this an option now.

To gain further insight into the behaviour of oxy-fuel flames at the shortest length scales, the experiments of chapter 7 could be repeated with a better laser that generates higher energy for the polarised/ depolarised Rayleigh measurement. This would increase the SNR for the temperature images, and allow a complete layer thickness analysis (including the thermal diffusivity term) to be performed. Furthermore, the scalar dissipation rate could be derived from high SNR mixture fraction images derived from the polarised/ depolarised measurement technique.
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