



Department of Mechanical Engineering
University College London

**Characterisation of Laser Induced Spark-Ignited Flame
Kernels in Premixed Fuel/Air Mixtures**

Emmanuel, Sunny Okara

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Philosophy (PhD) Award in Mechanical Engineering**

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Declaration

I, EMMANUEL Sunny Okara confirm that the report presented here is based on a personal effort. Where information has been derived from other sources, I confirm that this has been indicated in the report.

Abstract

Gaseous alternative fuels are promising solution for today's increasing demand for clean and reliable power. The wide number of fuel types and sources implies that engine designers need to develop fuel flexible combustors. Also, to meet tightening emission laws, these combustors would be required to operate under ultra-lean, high pressure and high temperature environments. Such extreme conditions make ignition difficult to achieve especially with current spark plugs which has been the primary ignition source during the last one hundred years. Laser ignition has been proposed as an alternative ignition system capable of providing stable combustion under these conditions. The advantages provided by laser ignition over electric spark system include: the absence of flame quenching effects of electrodes which enhances the ignition of lean mixtures, less energy requirement for ignition at higher pressures, precise timing, and choice of suitable ignition location. To explore the benefits offered by the Laser ignition in practical combustors, there is a need to characterise the propagation behaviour of the laser flame kernel since successful ignition requires the transition from an ignited spark kernel to a self-sustained flame.

The present thesis contributes to existing knowledge on laser ignition through investigation of different development characteristics of the ignited flame kernel. The first investigation involves high-speed imaging of the flow field characteristics of the flame kernel based on combined 2D Laser tomography and PIV techniques. The ignition was achieved by focussing a laser beam of 1064 nm wavelength on an atmospheric co-axial straight tube burner through which stoichiometric CH₄/Air was flowed. The resulting flame kernel and its flow field were visualized through laser-sheet illumination and then captured using a high-speed camera. The observed flame kernel features from the tomographic images were consistent with previous research observation and provided insight to other phenomena such as induced vortex motion in the developing kernel. Additionally, the PIV data provided insight on how the local flow field velocities were changing during development of the flame kernel.

The second investigation involves direct imaging of the flame kernel chemiluminescence to understand both the fluid dynamics and chemical reactivity of the laser flame kernel. The atmospheric burner used in this setup is made of co-axial contracting nozzles in which flowing fuel/air mixtures were repeatedly ignited by a focused laser beam of 1064 nm wavelength and 2 Hz frequency. To characterise the resulting flame kernel, 2D projection images of the kernel OH* chemiluminescence was captured using intensified CCD camera. The observed geometric features of the kernel were similar to the earlier observation. Additional data on the OH* luminosity provided insight on the reactivity of the kernel at various transition points during its development and the reason for the variation in growth rate of the flame kernel at different stages. The investigation was extended to include the effects of varying physical parameters such as laser pulse energy and flow velocity. The observation showed that the effect of increasing the pulse energy within a certain threshold limit is an enhanced early kernel growth, but the ultimate effect was insignificant. Although, a higher flow velocity had no remarkable effect on the size of the kernel, it resulted in faster propagation of the flame front downstream due to the combined effect of convection and increased turbulence.

In the final study, the sensitivity of the kernel characteristics to changes in the fuel thermochemical properties was investigated based on direct imaging of the OH* chemiluminescence. The investigation comprises the effect of changing equivalence ratio, variation in fuels at constant Adiabatic Flame Temperature and variation in fuels at

constant Laminar Flame Velocity. The results of the analysis showed linear dependence of most characteristics with equivalence in laminar flow regime but not in turbulent flows. For both constant temperature and constant laminar velocity mixtures, the results showed differences in the flame kernel characteristics depending on the fuel. This shows that no single thermochemical property is enough to uniquely define different fuel/air mixtures. Hence, further study on the inter-dependencies of the different thermochemical properties would be necessary for development of more robust model that would characterise flame kernel propagation in flexible combustion systems.

Thesis Impact

My PhD research has provided new set of data that could help explain how the choice of fuel and selection of appropriate ignition parameters can either improve or hinder stable combustion. These are important considerations that has direct impact on the emission of harmful substances from known combustion devices. The direct application of results from this work is in modelling premixed combustion process which is the combustion mode prevalent in most internal combustion engines as well as in accidental fire explosions.

As part of my PhD work, I developed two unique experiment measurement solutions which enabled me acquire data on the developing flame kernel geometry, the associated radical intensity level as well as the flow field velocity data. The first solution comprises a system of optical instruments combining Laser tomography and PIV techniques to enable simultaneous measurement of the laser-ignited flame kernel and its flow field in an atmospheric burner. The second solution involves a direct capture of the flame kernel using intensified CCD camera by filtering out radiations from the background. The selected techniques are well established and widely used in research.

Some results from this work have been presented locally in the annual UCL Mechanical Engineering PhD conference. My next plan of action is to present this work to wider audience by attending major international conferences and publishing papers. I am passionate about educating young students in primary and secondary schools about how they can impact their environment through knowledge of optics which is the science from which my research technique is based. While doing my PhD, I have been doing voluntary work with the STEM Ambassadors Network where I encourage students to take on STEM subjects. I have also been a member of The Combustion Institute UK and Institute of Physics (IOP) where I engage actively with fellow professionals in topics related to using science to solve emission problems.

Acknowledgement

I wish to thank my supervisors Prof. Alistair Greig and Prof. Rama Balachandran for their expertise guidance and direction through this research work. Prof. Greig, you supported me from onset when I brought my research idea, fine-tuned it to what it is today, and you did not give up on me even in the most difficult times. Prof. Balachandran, you inspired and directed my work, helping me with technical knowledge in the field and supported me with the platform to conduct my experiment.

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I am forever indebted to my family whose love, prayers and encouragement has kept me going. My wife, Ify has been my greatest source of strength during this period. My kids, Salem and Eunice, who were born during this period made my daily life to be fun and without a dull moment. My elder sister, Mrs. Blessing Youdiowei, who looked after me while growing up and has always been there for me. My siblings, Evans, Iya, Kate, Priscillia and Happi, who always go the extra mile in their support for me through sacrificial giving and relentless prayers. I also cherish the continuous love received from nephews, cousins, aunties, and in-laws.

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To my parents and role models, Mr. and Mrs. Emmanuel, who are now with the angels, I dedicate this work in your honour because you were the best dad and mum anyone could pray for. You will continue to live in my heart.

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Nomenclature

Symbols

α	Thermal diffusivity	m^2s^{-1}
c_p	specific heat of the mixture at constant pressure	$\text{kJkg}^{-1}\text{K}^{-1}$
D	mass diffusivity	m^2s^{-1}
δ_f	Flame thickness	m
ε	Dissipation rate of turbulent kinetic energy	m^2s^{-3}
E, E_p	Pulse energy	mJ
E_{min}	Minimum Ignition Energy	mJ
f	Pulse frequency	Hz
$h\nu$	photon energy	eV
l	Integral length scale	m
I	radiation intensity	AU
I_0	intensity threshold	AU
k	Thermal conductivity	$\text{Wm}^{-1}\text{K}^{-1}$
K	Stretch rate	$1/\text{s}$
L	characteristic length	m/s
ℓ	Markstein length	m
Le	Lewis Number	
λ	Laser wavelength	nm
N_e	electron density	m^{-3}
$N_{e,0}$	starting electron density	m^{-3}
η	Kolmogorov length scale	m
\emptyset, φ	Mixture Equivalence ratio	
ρ	fluid density	kg/m^3
ρ_u	Unburnt gas density	Kg/m^3

ρ_b	Burnt gas density	Kg/m ³
Q	heat released per unit mass fraction of the deficient reactant	kJ/kg
Re	<i>Reynolds Number</i>	
S_{y+}	Flame front propagation rate along $Y_+ \left[\frac{dY_+}{dt} \right]$	m/s
S_{y-}	Flame front propagation rate along $Y_- \left[\frac{dY_-}{dt} \right]$	m/s
S_{x+}	Flame front propagation rate along $X_+ \left[\frac{dX_+}{dt} \right]$	m/s
S_{x-}	Flame front propagation rate along $X_- \left[\frac{dX_-}{dt} \right]$	m/s
S_L	Laminar flame propagation rate	m/s
S_f	Stretched flame speed	m/s
T_{ad}	Adiabatic flame temperature	K
T_u	unburned gas temperature	K
T_f	Flame temperature	K
T_g	Gas temperature	K
τ	Integral time scale	s
τ_e	time constant	s
U	mean velocity	m/s
u	instantaneous velocity	m/s
\bar{u}	mean velocity	m/s
u'	fluctuating velocity	m/s
u_L	Unstretched laminar flame speed	m/s
U_0	Bulk flow velocity	m/s
μ	dynamic viscosity	m/s
v_d	diffusion rate of electrons	s ⁻¹
X	Flame kernel width	mm
X_+	flame tip location away from Laser source	mm
X_-	flame tip location towards the Laser source	mm
x_a	mole fraction of the air	
x_f	mole fraction of the fuel	
Y	Flame kernel height	mm
Y_t, Y_+	Downstream flame tip location	mm

Y_b, Y_-	Upstream flame tip location	mm
Y_u	mass fraction of the deficient reactant	

Abbreviations

AFT	Adiabatic flame temperature
CH*	CH radical
CH ₄	Methane
C ₃ H ₈	Propane
C ₂ H ₄	Ethylene
CCD	Charged Coupled Device
CNG	Compressed Natural Gas
FF	Flame front
FFT	Fast Fourier Transform
IE	Ionization Energy
LFL	Lean flammability limit
LFV	Laminar flame velocity
LPG	Liquified Petroleum Gas
LNG	Liquified Natural Gas
LI	Laser ignition
LISI	Laser induced spark ignition
MARPOL	Maritime Pollution
MIE	Minimum Ignition Energy
MPE	Minimum Pulse Energy
MW	Mega watts
NG	Natural Gas
OH*	Hydroxyl radical
PIV	Particle Image Velocimetry
PLIF	Planar Laser Induced Fluorescence
TL	Threshold Level
TTL	Transistor–transistor logic

UFL Upper flammability limit

UV Ultraviolet

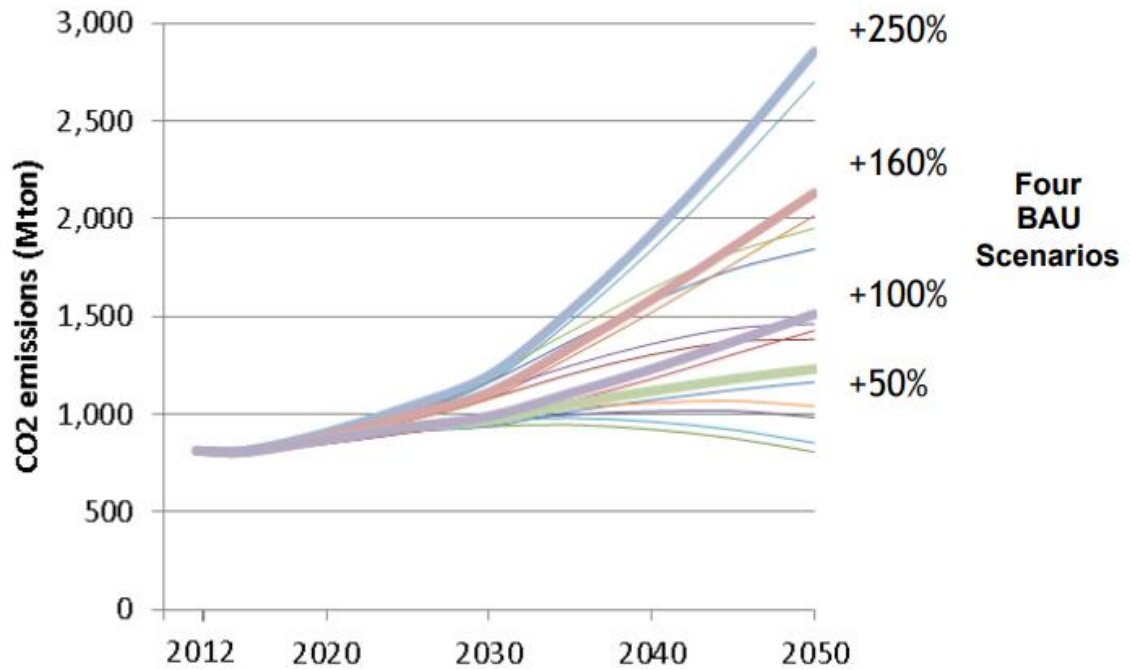
1

Chapter 1 Introduction

2 1.1 Thesis Background

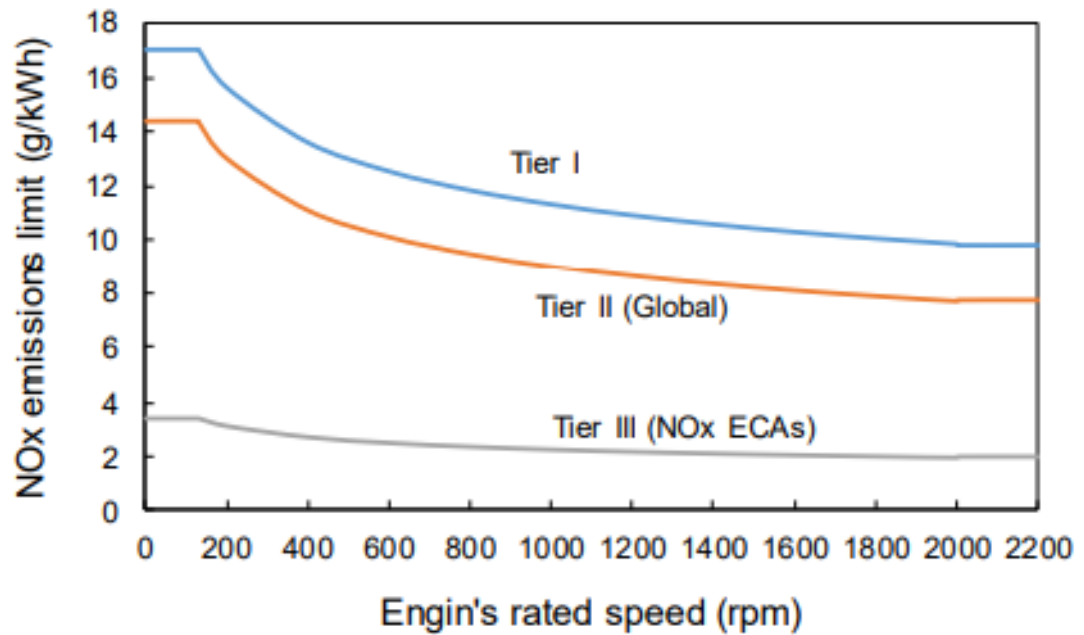
3 1.1.1 The Global emissions challenge.

4 The global community is threatened by the effects of rising emissions from the
5 combustion of fossil fuels used in the transport and energy sector. A 2009 study by
6 the International Maritime Organisation [1], showed a rising annual trend in
7 shipping emission of CO₂ and other greenhouse gases (GHGs). As shown in Figure
8 1.1, projections reveal that future CO₂ emissions could rise between 50% and
9 250% from 2012 to 2050 depending on what the future economic and energy
10 developments scenario will look like [2]. This increased concentration of CO₂ and
11 other GHGs is the main cause of global warming, and the resulting effects includes
12 loss of ice cover near the poles, global average rise in sea levels, etc. In addition to
13 global warming effects, the burning of fossil fuels in most IC engines pose a threat
14 to human health due to the release of poisonous gases like unburnt hydrocarbons
15 (HCs), oxides of nitrogen (NO_x), oxides of sulphur (SO_x) and carbon monoxide
16 (CO). Hydrocarbons are harmful to humans, affecting internal organs.
17 Hydrocarbons together with oxides of nitrogen form ground level ozone, which is
18 the primary component of smog [3]. Carbon monoxide is very dangerous, with
19 human exposure to high levels being fatal.



1
2 **Figure 1.1: Projection of CO₂ emission under four Business-As-Usual (BAU)**
3 **Scenarios [2]**

4
5 As counter measures to these threats, stringent emission regulations have been
6 imposed in different sectors. In the maritime industry, the MARPOL Annex VI set
7 limits on NO_x and SO_x emissions from ship exhausts and prohibits deliberate
8 emissions of ozone-depleting substances. Figure 1.2 shows the NO_x emission limits
9 for different ship categories which depends on the vessel's construction date and
10 engine rated speed. According to this regulation, ships constructed before 1
11 January 2000 installed with diesel engines above 5000kW are required to comply
12 with Tier I standards while ships built after 2015 must comply with either Tier III
13 or Tier II standards depending on the zone of operation. Another legislation aimed
14 at combating pollutant emissions from SI engines is the European Passenger Car
15 Emissions Legislation for Gasoline IC engines (Table 1.1) originally specified in
16 Directive 70/220/EEC which was later replaced by Regulation 715/2007 (Euro
17 5/6). Both legislations trends show an increasing strictness on emissions due to
18 growing environmental concerns, highlighting the need for alternative fuel sources
19 and development of advanced ignition technologies for IC combustion engines.



1

2 **Figure 1.2: NOx emissions regulations in marine shipping based on rated engine**
 3 **speed [10]**

4

5 **Table 1.1: European Passenger Car Emissions Legislation for Gasoline IC engines**
 6 **[4]**

	Year	CO	HC	NOx
		<i>g/km</i>		
Euro IV	2005	1.0	0.10	0.08
Euro V	2009	1.0	0.10	0.06
Euro VI	2014	1.0	0.10	0.06

7

8

9 **1.1.2 Gaseous fuel solution and its challenges**

10 One of the temporary measures being used to cope with the challenges of
 11 emissions and regulations is by shifting to gaseous fuels. Although these fuels have
 12 established usage in sectors like power generation and aviation, their popularity is
 13 beginning to gain ground in the automobile and marine sectors. The advantage of
 14 gas-phase fuel over liquid-phase fuel is that they have high combustion efficiency
 15 over wide firing ranges. This is due to their ability to form homogenous fuel-air

1 mixtures in less time. They also form cleaner combustion products with hardly any
2 solid pollutants (e.g., ash, particulates, or soot) compared to liquid-phase fuels.

3 Natural gas (NG) is one of the promising gaseous fuels with cleaner combustion
4 products compared to other commercially available liquid fuels. The number of
5 engines powered by NG has been growing rapidly in both automobile [5] and
6 marine sectors [6]. Figure 1.3 is a recent DNV report which shows that there
7 currently 247 confirmed LNG fuelled ships with additional 110 ready ships.
8 Methane, which is the main component of NG (~ 90% CH₄) stands out as an ideal
9 experimental fuel for demonstrating ignition performance of gaseous fuel sources
10 because of its unique properties. For example, due to its large C-H bond energies,
11 methane has a relatively high ignition temperature and low laminar flame speed.
12 In addition, it has unity Lewis number which means its data can be easily applied
13 to modelling validation by neglecting Lewis number effects.

14 Liquefied Petroleum Gas (LPG), a product of petroleum refining is a feasible
15 alternative to petrol which could lower CO₂ and other pollutant emissions in SI
16 engines. Propane which is one of the liquefied petroleum gases (LPG) is a readily
17 available and adaptable engine fuel which can be liquefied and stored at low
18 pressures; hence its transportation is more convenient than other gaseous fuels.
19 Like other LPGs (e.g., butane), propane has relatively high calorific value and
20 octane number. Its use as an alternative transport fuel is rising with about 17
21 million vehicles worldwide [7].

22 Ethylene which is a by-product of steam cracking of longer chain hydrocarbons (an
23 energy intensive process that involved temperature of between 750-950 degrees
24 Celsius) has thermo-physical properties comparable to gasoline which are strong
25 indications that the gas can be easily combusted. For example, both the enthalpy of
26 combustion (49.46 MJ/kg) and autoignition temperature (490°C) indicates better
27 combustibility of the gaseous fuel in conventional gasoline in SI engines[8]. It is
28 commonly used in agriculture to enhance the ripening of fruits and to stimulate
29 rubber tree to yield more latex. Also used in petrochemical industry for polythene
30 production by polymerization. In 2005, the global production ethylene exceeded
31 75 million metric tonnes per year [8].

1 The present study includes an investigation of the combustion of three
2 hydrocarbon fuels (methane, propane, and ethylene) with unique properties. One
3 issue of concern with the wide number of fuel types and sources is the impact of
4 fuel composition variation on flame stability of combustors. Combustion instability
5 due to fuel variation is caused by differences in the laminar burning velocities (S_L)
6 and adiabatic flame temperatures (T_{ad}) of the fuels. Therefore, the impact of fuel
7 variability on combustion needs to be looked at to minimise the impact of engine
8 pollutant emissions.

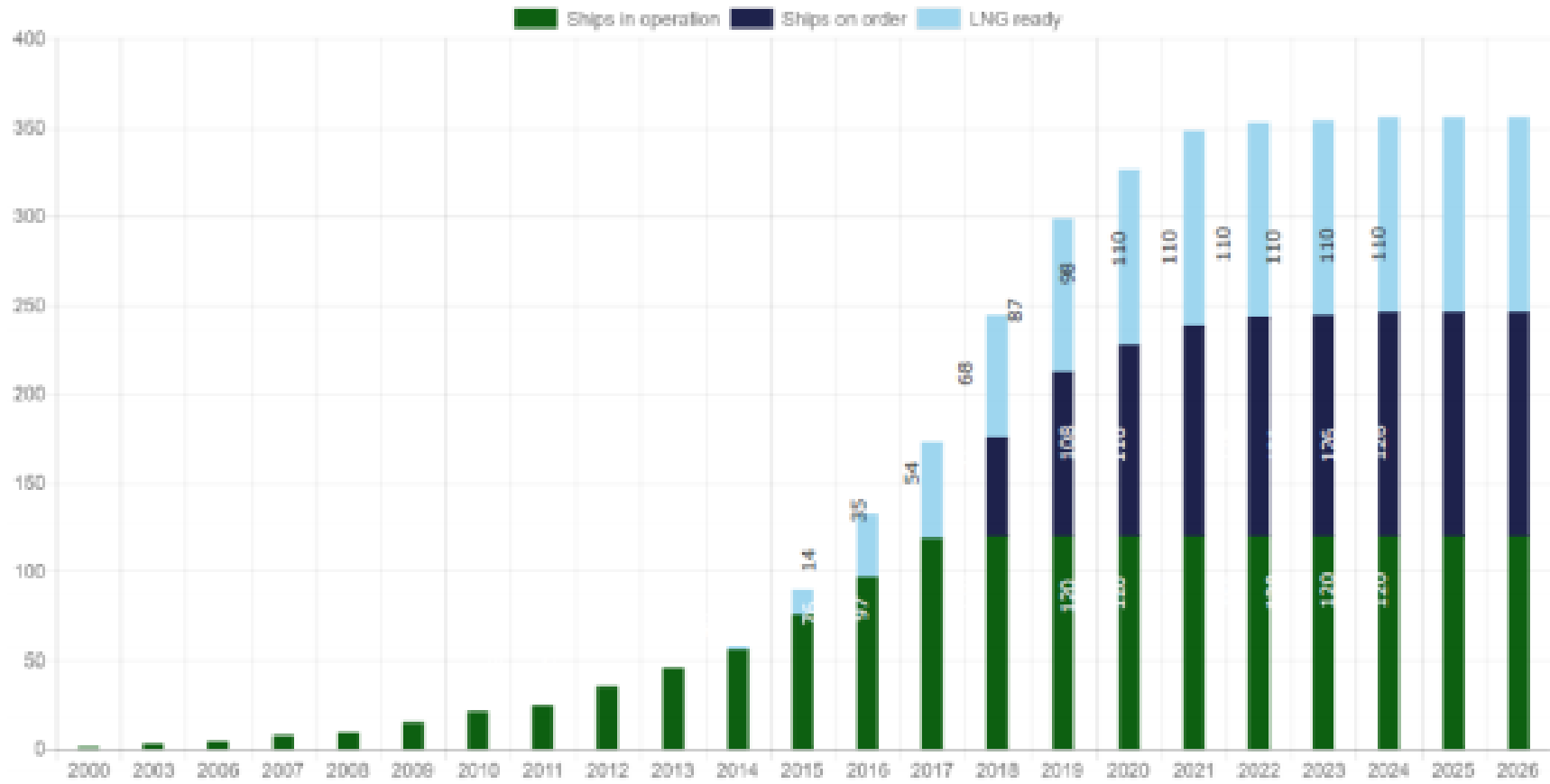


Figure 1.3: Existing and planned LNG Fleet [6]

1

2

1 1.1.3 Lean premixed operation and the challenges with spark plugs

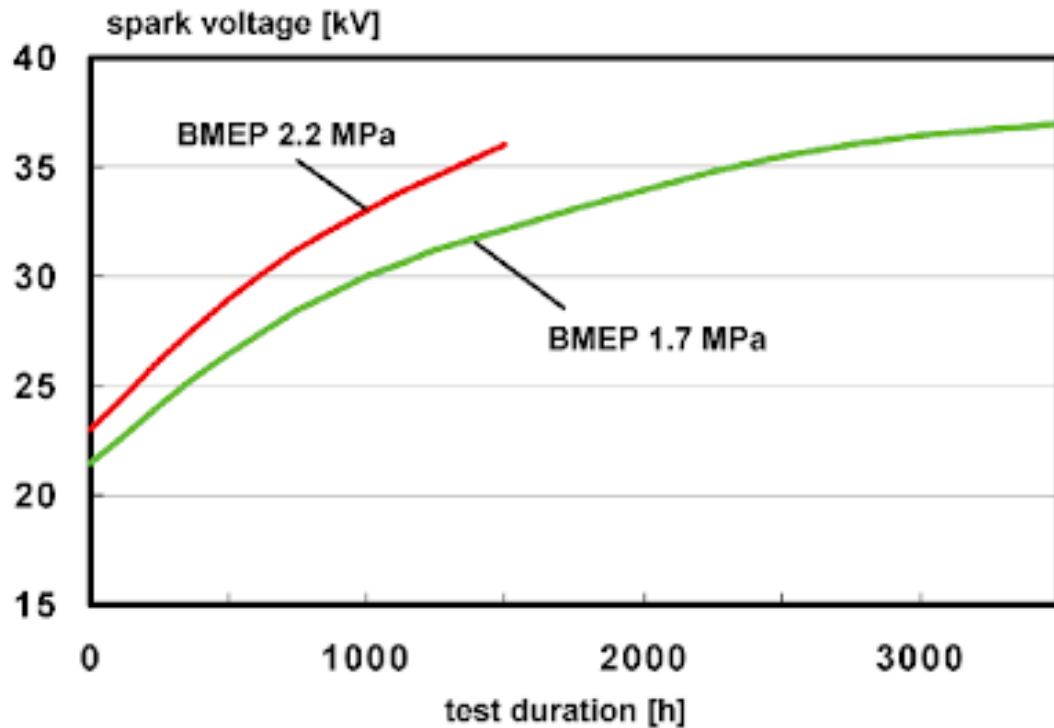
2 Emissions from gas-fuelled engines are further improved by lean premixed
3 operation [9], [10]. By igniting with leaner fuel-air mixtures, NO_x emissions are
4 reduced due to the combustion temperatures being lowered, however this leads to
5 considerable reduction in power density of the engine. To compensate for this,
6 lean mixture combustion requires higher compression ratios (CR) as shown by
7 Equation 1.2 [11].

$$8 \quad \eta = 1 - \frac{1}{(CR)^{\gamma-1}} \quad (1.2)$$

9 γ = the gas adiabatic coefficient ($\gamma = \frac{c_p}{c_v}$) which is related to the leanness or
10 richness of the fuel/air mixture.

11 The resulting higher efficiency however comes at a cost due to reduced lifespan of
12 ignition spark plugs. Figure 1.4 shows the required voltage for ignition as a
13 function of spark plug lifetime for brake mean effective pressures of 22 bar and 17
14 bar in a gas engine. As shown by the figure, a higher spark plug voltage would be
15 required for ignition which significantly reduces the lifespan of spark plugs. This
16 trend can be explained by Paschen's law [12] which states that the breakdown
17 voltage (V) for a uniform electric field at constant temperature is a function of the
18 product of the gas pressure and the spark gap [i.e. $V = f(p, d)$].

19 The above situation is compounded by the push for more stringent emissions
20 regulation form the basis for research towards the development of alternative
21 ignition sources to the spark plug system which has been the main ignition sources
22 for combustion engines in the past one hundred years. To tackle this problem, a
23 number of novel ignition concepts like high-energy spark plugs, plasma jet igniters,
24 rail plug igniters, flame jet igniters, torch jet igniters, pulsed-jet combustion and
25 laser ignition systems are currently under development [13], [14]. In the current
26 thesis, the focus is on laser ignition which has been proposed as viable alternative
27 to spark plug system capable of providing stable combustion under extreme
28 environmental conditions.



1

2 **Figure 1.4: Breakdown Voltage of Gas engine Spark Plugs at two BMEP levels [15]**3 **1.1.4 Laser ignition and its benefits**

4 An alternative ignition source for gas-fuelled engines is the use of a laser beam for
5 ignition. Laser ignition involves focusing a laser beam to a small volume of fuel-air
6 mixture until the peak intensity of the focal volume exceeds a certain threshold
7 (known as the breakdown intensity threshold). When this happens, a breakdown
8 of the medium occurs leading to the formation of a spark plasma, whose size
9 depends on the Numerical Aperture of the focused laser beam. If the energy
10 content of the spark plasma is higher than a certain value (known as the Minimum
11 Ignition Energy), the mixture ignites which can result in the propagation of a self-
12 sustained flame. This process differs from spark ignition in many ways. For
13 example, the ignition time scale is much shorter, and the breakdown process is
14 photon driven unlike dielectric breakdown obtainable with spark electrodes.

15 Ronney [10] discussed four possible ways a laser can ignite a flammable mixture:
16 thermal initiation, non-resonant breakdown, resonant breakdown and
17 photochemical ignition. Thermal initiation (TI) involves using a laser source to
18 heat up a flammable mixture beyond its threshold ignition temperature [16]. The

1 method is most suitable for fuel/oxidizer mixtures with strong absorption at the
2 laser wavelength, but it involves long ignition delay times [14]. Photochemical
3 ignition (PCI) involves use of laser energy to dissociate target molecules within the
4 mixture into highly reactive radical species, leading to ignition[14], [17]. The
5 method requires less energy, and the mixtures can be ignited at lower pressures
6 and closer to their flammability limits. However, it requires a close match between
7 the laser excitation wavelength and the target molecules absorption wavelength;
8 hence a tunable laser is required. Resonant breakdown ignition (RB) process
9 involves, first, a non-resonant multiphoton dissociation of molecules resulting in
10 free atoms, followed by a resonant photoionization of these atoms[18], [19]. This
11 process generates sufficient electrons needed for gas breakdown. Theoretically,
12 less input energy is required due to the resonant nature of this method. Of all the
13 mechanisms, non-resonant breakdown ignition (NRB) is the most feasible in terms
14 of practical implementation. It involves using a focused laser beam to create an
15 electric field of sufficient intensity within a flammable mixture thereby leading to
16 breakdown of the medium[10], [14]. Unlike PCI and RB, it does not require a close
17 match between the laser wavelength and the absorption wavelength of target
18 molecules, hence it can easily be implemented in practical combustion
19 systems[14]. It is also the most similar to the electric spark ignition involving
20 production of a spark plasma which emits light, heat and shockwave; hence, it is
21 also called Laser Induced Spark Ignition (LISI).

22 There are several potential benefits which laser ignition systems provide over
23 electric spark plugs due to differences in their physical configurations. For
24 example, lasers are non-intrusive which eliminates problems associated with the
25 presence of electrodes which act like heat sink and tend to quench the flame kernel
26 especially in lean mixtures. Due to its flexible nature, laser ignition can be achieved
27 with precise timing and choice of suitable ignition location. Additionally, the
28 differences in plasma parameters (initial temperature, pressure, electron
29 parameters, etc.) of both systems results in differences in their performance. For
30 example, in contrast to electric sparks which is governed by Paschen's law, laser
31 ignition requires lower energy at higher pressures. Previous studies also found
32 that the laser ignited flame speed at early times, exceed the laminar flame speed,
33 thereby providing a clear indication of plasma-assisted flame propagation[20].

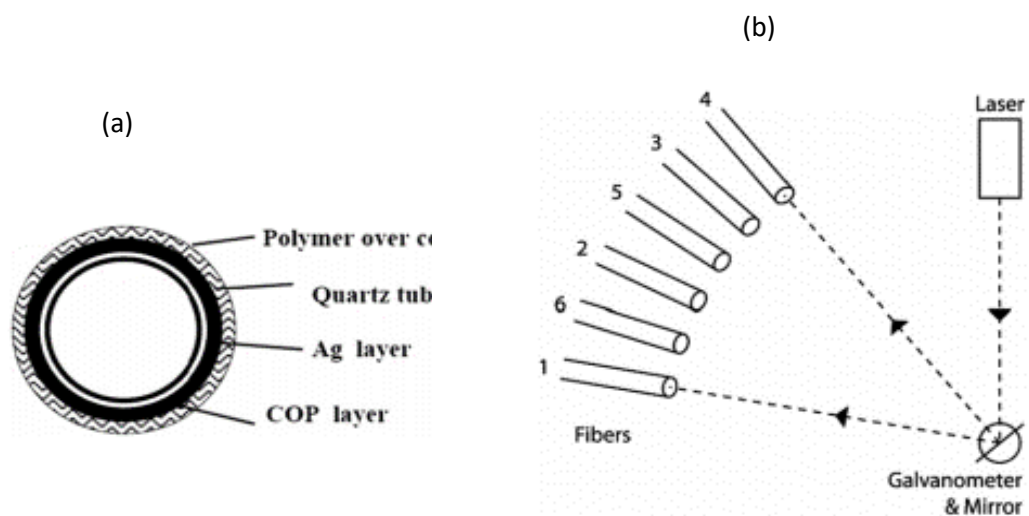
1 Due to its benefits, the implementation of laser ignition is highly sought in a host of
2 combustion applications including: stationary engines [21], [22], internal
3 combustion engine [23], aircraft engines and rocket engines [24]. In stationary gas
4 engines, high cyclic variation which results from slow burning of lean mixtures can
5 be reduced via multipoint ignition or choosing an appropriate ignition location. It
6 is also known that unlike the electric spark, the dependence of laser ignition
7 energy on flow pressure is the opposite of Paschen's law, which means less energy
8 is required for ignition. In spray-guided direct injection Otto-engine, where charge
9 stratification is critical for achieving fuel economy at low loads, optimization of the
10 ignition process is extremely difficult due to fixed spark plug location. Laser
11 ignition makes this easy through flexible spark location. In addition, the formation
12 of the spark plasma is adaptive with respect to the occurrence of fuel droplets near
13 the focus because condensed matter reveals lower breakdown thresholds than
14 gases ($\sim 10^9$ vs. 10^{11} W/cm², respectively) [25],[23]. In homogenous charge
15 compression ignition engine, the self-starting reactions with a high temporal jitter
16 is a common problem that severely affects the smooth running of the engine. Laser
17 ignition allows triggering of the start of "self-ignition" by introducing nearly
18 negligible energy into the cylinder with optimum timing. In this way it can help
19 substantially to overcome such drawbacks [26], [27]. In aircraft gas turbine, high
20 altitude flame extinction is a common problem which can be caused by transient
21 disturbances of the air flow through the engine, or by severe ingestion of ice,
22 water, and dust [28]. When the engine is extinguished, the temperature and
23 pressure in the combustor are low, causing a significant decrease in vapour
24 pressure of fuel and the decreased air flow rate may lead to poor atomization, both
25 of which imply the need for large amount of spark energy to initiate a flame kernel.
26 Therefore, there is rising interest in using laser plasmas for ignition of turbines
27 used in aircraft engines primarily in order to achieve rapid relight [29], [30], to
28 capitalize on the possibility of more optimal spark locations along the centreline of
29 the combustor or in flow reversal zones near the fuel nozzle [31], and to avoid the
30 reliability limitations of conventional igniters.

1 1.1.5 Practical implementation of laser ignition and associated difficulties

2 Despite the benefits offered by laser ignition, the concept is yet to be adopted in
3 most practical combustion systems. One drawback towards the practical
4 implementation of laser ignition is the huge cost and size of equipment needed to
5 generate high power pulsed laser beams. From inception of laser ignition research,
6 conventional flash-pumped solid-state lasers (e.g. neodymium-doped yttrium
7 aluminium garnet or Nd:YAG lasers) have been used to generate fundamental
8 lasing frequencies in the near infrared region. This fundamental frequency can be
9 doubled to produce visible beam, and further doubling can produce ultraviolet
10 beam. In recent decades, considerable progress has been made in laser technology
11 to reduce the size and cost of potential lasers for ignition much of which stems
12 from improvements to laser diodes and diode pumped solid state lasers. Research
13 efforts has shown that it is now possible to create diode pumped solid state micro-
14 lasers with an energy output of several millijoules; enough to cause laser-induced
15 breakdown [32],[33]. Some researchers have suggested that the use of diode
16 pumped laser sources has advantages over flashlamp pumped lasers, which
17 usually result in variation in the output beam properties across the operating
18 range, limitation of beam repetition frequencies and a shorter life cycle [34],[35].

19 The most common research technique employed in the initiation of laser ignition is
20 open-path beam delivery which uses mirrors and lenses to create gas breakdown
21 within the combustion location. Such configurations may work over short
22 distances; however they are not practical for commercial implementation owing
23 safety, maintenance, thermal and vibrational issues which can cause misalignment
24 problems [14]. There are two promising concepts by which the commercial
25 delivery of a laser beam into the combustion chamber can be realized, namely the
26 laser mounted on the cylinder or located at a remote location [27]. Both concepts
27 are possible through adoption of optical fibre delivery which could be used to
28 transport either the ignition pulse or pump pulse. Yalin et al. [36], [37] was the
29 first to demonstrate ignition in a natural gas engine using a hollow fibre optic cable
30 (Figure 1.5a) coupled to a remote flash lamp pumped Nd:YAG laser source. The
31 tests showed that the hollow fibre delivery system met the goal of providing a
32 robust and successful means of engine ignition. Engine data showed that the fibre

1 coupled system yielded 100% reliability in ignition and reduced combustion
 2 durations compared to conventionally ignited cylinders [38]. A potential advantage
 3 of the hollow fibre spark delivery approach is the ability to use a single laser
 4 source multiplexed via multiple fibres (Figure 1.5b) to a series of engine cylinders.
 5 Thus, the development of a flexible optical fibre system for practical
 6 implementation in internal combustion engines is highly desirable. However, it can
 7 be quite challenging to bring laser ignition into a realistic application due to the
 8 high peak power required to generate laser sparks, however, there are many
 9 research efforts which focussed on addressing this problem [39], [29].



10
 11 **Figure 1.5: Fibre optic beam delivery: (a) cross-sectional view of hollow core**
 12 **coated fibre, (b) Step and hold multiplexing employing mirror attached to**
 13 **galvanometer [14].**

14 1.1.6 Important Characteristics of the Flame Kernel

16 The processes involved in laser ignition are very complex, therefore optical
 17 diagnostics are usually employed by researchers to gain insight into the key
 18 events. The choice of a particular technique is based on what information is of
 19 interest to the researcher. For example, while some techniques (e.g., shadowgraph,
 20 schlieren) provide information related to the fluid dynamics of the flame kernel
 21 others (e.g., chemiluminescence, PLIF) provide additional details of the chemical

1 kinetics. In this research, both the fluid dynamics and chemical kinetics data of
2 interest so as to understand the coupling between the two.

3 In IC engines, geometric properties of the flame kernel such as the shape and size
4 are often investigated. These are useful for predicting the pressure rise with time
5 which maybe too small to be detected by existing pressure probes [40],[41]. They
6 are also useful for estimating the *burning velocity* which is a fundamental
7 parameter required for the development and validation of kinetic mechanisms for
8 fuels. In the early stage of laser ignition, such measurements are particularly
9 important because the relative expansion speed of the flame kernel is low
10 compared with that of a fully developed flame. Also, the laser flame kernel is
11 subjected to high stretch rates which makes extinction more likely. For gaseous
12 fuels, the risk of quenching is high due to its lower energy density compared to
13 liquid-based fuels, hence the need to understand the coupling between the kernel
14 physical properties and ignition success. In addition, there is the widely held view
15 that most practical ignition problems (e.g. quenching, slow burning, unstable
16 combustion, emissions, etc.) originates at this stage [42],[14],[43].

17 Another important characteristic of the early flame kernel is the rate of radical
18 generation at the kernel-fluid boundary which determines whether the kernel
19 propagates further or extinguishes. Excited radicals are formed in the flame by
20 thermal excitation and through chemical reactions, with concentrations much
21 higher than their equilibrium values. The main chemiluminescence emitters in
22 hydrocarbon flames are OH*, CH*, C₂* and CO₂* [44], [45]. Their formation
23 reactions are detailed in Table 1.2, together with the band wavelengths. As shown
24 in the table, these radicals are formed in reactions involving intermediate
25 combustion species, whose concentration in the flame exceed their equilibrium
26 values. Therefore, since de-excitation reactions are proportional to the
27 concentration of the excited radicals and have very short time, chemiluminescence
28 is mainly originated in thin reaction zones. This can be observed in Figure 1.6,
29 representing the spatial distribution of local emission intensities in a laminar
30 premixed flame. The reactions listed in the table strongly depend on temperature
31 and involve stable and intermediate fuel and oxidizing species. All of them are
32 important combustion parameters and, therefore, the resultant

1 chemiluminescence emission might be expected to be closely related to the
2 properties of the flame. In fact, spontaneous flame radiation has been extensively
3 used as a flame diagnostic method.

4 In the current study, the chemiluminescence of OH* will be used as flame markers
5 to detect flame position, shape, and structure, while its intensity is used to indicate
6 the relative heat release rate. The use of OH* chemiluminescence as flame marker
7 has been demonstrated in many past studies [46],[47],[48]. Also, there are many
8 research evidence of the connection between the chemiluminescence intensity of
9 OH* and equivalence ratio [49],[50],[51]. According to [52], the chemical reactions
10 occurring during an ignition event involves three key steps (chain initiating, chain
11 propagation, and chain termination). Initiation of the chemical reactions usually
12 occurs in a few hundred microseconds after the breakdown. The next step which is
13 the most important is the chain propagating step usually involves production of
14 OH* [52]. Therefore, OH* being a key radical produced during the chain
15 propagating step is an important ignition parameter and a reliable flame marker.

16 At present, direct measurement of HRR is not practical instead the production rate
17 of certain radicals (e.g. OH* and CH*) are used to indicate HRR [53],[54],[55]. A few
18 works have addressed specifically the existence of a relationship between
19 chemiluminescence and heat release rate. One of the earliest studies is that of Price
20 et al. [56], showing a linear relationship between mean C₂* emission and volume
21 flow rate of combustible, not influenced by the conditions of turbulence. Lawn [57]
22 evaluated the spatial cross-correlation of chemiluminescent emissions and
23 concluded that it might serve as a good indicator of instantaneous heat release
24 rate. Comparison of spatially resolved measurements of OH* and CH*
25 chemiluminescence, flame surface density and heat release rate estimated as the
26 product [CH₂O]_x[OH] revealed similar patterns and behaviour and, therefore, that
27 either OH* emission or flame surface density serve to estimate heat release rate
28 [58], [59]. Measured trends of OH*, CH* and CO₂* chemiluminescence intensity in
29 [60], [61] for variations in equivalence ratio and strain rate suggest that these are
30 good markers for heat release rate whereas C₂* is not a reliable indication.
31 However, Nori and Seitzman [62], [63] noted that due to the influence of
32 equivalence ratio and pressure on OH* and CH* emission, their associated signals

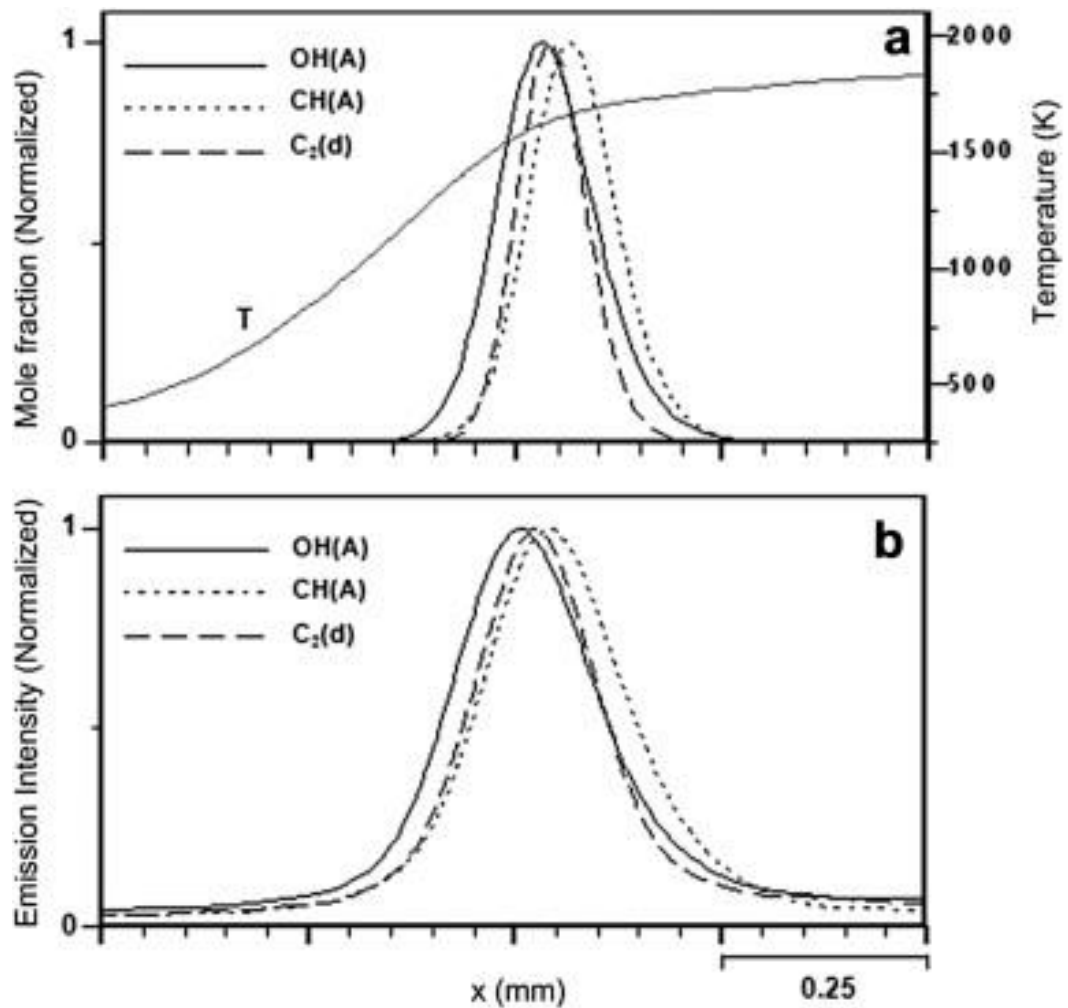
1 may not be fully reliable as heat release markers. Numerical results of Najm et al.
 2 [64] suggest that chemiluminescence due to OH*, C₂* and CH* may fail as local
 3 markers of heat release in high curvature regions of flames; hence the need for
 4 additional work and experimental evidences in order to verify that conclusion. For
 5 the case of internal combustion engines, Kim et al. [65] found a good correlation
 6 between chemiluminescence in the range 350–390 nm and heat release rate in
 7 cool flames.

8

9 **Table 1.2: Formation routes of excited radicals and characteristic wavelengths[66].**

Radical	Reactions	Wavelength (nm)
OH*	R1: CH + O ₂ → CO + OH* R2: H + O + M → OH*+M R3: OH + OH + H → OH*+H ₂ O	282.9, 308.9
CH*	R4: C ₂ H + O ₂ → CO ₂ + CH* R5: C ₂ H + O → CO + CH*	387.1, 431.4
C ₂ *	R6: CH ₂ + C → C ₂ * + H ₂	513, 516.5
CO ₂ *	R7: CO + O + M → CO ₂ * + M	350–600 (Continuous spectrum)

10



1

2 **Figure 1.6: Intensity profiles of OH* (306 ± 7 nm), CH* (431.4 ± 0.75 nm) and C₂* (516.5 ± 1 nm) chemiluminescence in a laminar methane-air premixed flame ($\phi =$**
 3 **1.1) along the flame normal direction. (a) Simulation (b) Experiment [66]**
 4

5

6 **1.1.7 Survey of Relevant Literature.**

7 The history of Laser Ignition research can be traced back to the time of first
 8 discovery of laser-induced optical breakdown by R.W. Terhune and co [67] where
 9 a beam of a pulsed Q-switched ruby laser of tens of MW peak power, was focused
 10 in air by a single lens to create a spark comparable to an electrical discharge
 11 between electrodes. This experimental set-up was dubbed 'the most expensive
 12 Spark Plug in automotive history' by Terhune which may have contributed to many
 13 researchers abandoning the research due to the belief that it would never be
 14 economically feasible. However, the last three decades has witnessed renewed

1 interest in laser ignition research. This no doubt may have been driven by
2 advances in laser technology as well as the current trends towards ultra-lean
3 combustion [68].

4 Although not exhaustive, the following literature publications deals with laser
5 ignition investigation which relevant to the present study. Early work by Bradley
6 et al. [20] and Phuoc [69] provide extensive reviews on the breakdown and spark
7 evolution process. Reviews by Tauer et al. [27] and Morsy [14] also provided
8 further information on the application of laser-induced ignition and current state
9 of art.

10 **1.1.7.1 Laser induced breakdown and plasma studies.**

11 A Laser Induced Spark Ignition (LISI) event usually involve three development
12 phases (spark generation, development of ignited flame kernel and propagation of
13 self-sustained flame), where failure of any one phase results in ignition failure. The
14 first phase which is the spark generation phase involves focusing a laser beam onto
15 a small volume of fuel/air mixture until the peak intensity of the focal volume
16 exceeds the breakdown intensity threshold of the medium, thus leading to gas
17 breakdown. Growth of the spark is accompanied with shock waves which leads to
18 loss of spark energy.

19 Following the development of lasers in the early 1960s, initial research effort was
20 focused on the parameters affecting this breakdown threshold. One of the earliest
21 studies by Mitsuk et al. [70] was the effect of focal length and pressure on
22 breakdown threshold of xenon and krypton at low pressures (0.06 - 0.75 atm).
23 They found that the threshold field intensity strongly depends on the focal distance
24 for low pressures where plasma diffusion losses are present.

25 A later study by Chan et al. [71] investigated the effects of focal length and
26 pressure for an additional number of gases, showing a similar trend.

27 Of importance to combustion system applications is how the breakdown threshold
28 varies in fuel/air mixtures of various compositions. Hickling and Smith [72]
29 studied the breakdown energy of various combustible fuels (isooctane,
30 cyclohexane, n-heptane, n-hexane, clear-indolene, and No. 1 diesel) in air mixtures.

1 They reported no significant differences between the breakdown energy of air and
2 the energies of the fuels studied.

3 Morgan [16] presented a theoretical and experimental study of the processes
4 involved during laser-induced breakdown in air. He identified the necessity of
5 using high spatial and temporal resolution to capture the phenomena, in order to
6 characterize the processes leading to the breakdown and presented a partially
7 unified theory to explain the phenomena in air. The process is described as a
8 competition between multi-photon absorption and inverse Bremsstrahlung with
9 deionization by diffusion, recombination, and radiative energy losses.

10 YalKcin et al. [73] examined the influence of ambient conditions on the laser air
11 spark and studied laser-induced breakdown under different conditions (humidity,
12 laser energy, particulate level, etc.). They found that the laser spark characteristics
13 were not very sensitive to the ambient conditions or the laser energy. This is
14 encouraging for the quantitative use of laser-induced breakdown spectroscopy
15 (LIBS) and is also of interest when applying laser-induced breakdown for ignition.
16 They found a good correlation between the radiative expansion of the laser spark,
17 using results obtained from a laser-supported radiation wave model. LIBS is an
18 optical diagnostic technique widely used in quantifying the atomic and molecular
19 species present in gases, liquids and solids details of which can be found in [74],
20 [75].

21 Minowa et al. [76] investigated the earliest stages of laser-induced air optical
22 breakdown using the Mie scattering theory and three different wavelengths: 1064,
23 532, and 355 nm. These results were in good agreement with the plasma that
24 forms with highly ionized plasma balls.

25 Dors et al. [77] analysed the fluid dynamic effects following laser-induced optical
26 breakdown. These authors identified the importance of the plasma shape on the
27 shock wave formation. The shock wave formation process is of first-order
28 importance to explain the toroidal shape observed after the plasma cooling phase.

29 Although this is outside the scope of the present study, understanding the laser
30 plasma development dynamics will help explain some of the characteristic features
31 of the flame kernel.

1 1.1.7.2 Ignition studies: Measurement of Minimum Ignition Energy (MIE).

2 Many experimental studies on laser ignition involved measurement of the
3 Minimum Ignition Energy (MIE). Growth of the spark is always accompanied with
4 shock waves which leads to loss of energy. A means to investigate ignition success
5 during the plasma development stage is through measurement of the spark energy
6 which must be higher than the MIE of the combustible mixture. In practical terms,
7 a minimum pulse energy (MPE) greater than the MIE must be supplied by the laser
8 source to achieve successful ignition.

9 One of the first measurements of minimum ignition energy (MIE) of laser ignited
10 methane-air mixtures was by Weinberg & Wilson [78] using a ruby laser of pulse
11 energies of 2 J and duration 20 ns full width at half maximum (FWHM). Here, the
12 pulse energy was measured by focusing the laser beam through a small aperture
13 into an absorbing spherical calorimeter. For the first time, it was shown that MIE
14 and the plasma dimensions decreased with increasing pressure.

15 Kingdon & Weinberg [79] analysed the effect of pulse duration and plasma
16 constitution on MIE where they found out that for short pulse duration, the MIE
17 was independent of plasma constitution, while for longer duration pulses (1 ms)
18 the presence of inhibitors in the plasma could lead to flame extinction.

19 Syage et al. [80] measured the ignition energy of Hydrogen/air mixtures of
20 different equivalence ratios using output at 1064, 532, and 355 nm of a Nd-YAG
21 laser operating either as a Q-switched nanosecond laser or a pulse-mode-locked
22 picosecond laser. They reported minimum ignition energies that are higher than
23 the electric-discharge ignition energies and that increase toward the fuel-lean and
24 rich side of the stoichiometry. Also, MIE dependence on gas composition and
25 equivalence ratio for different combustible gases have been measured by both
26 Phuoc [81] and Beduneau et al. [81], [82]

27 Other published data on MIE by Huang et al. [83], Shy et al. [79] and Cardin et al.
28 [85] reveal the occurrence of key ignition transition events (in terms of increasing
29 MIE) for different experimental conditions.

30 There are many other experimental studies based on measurement of MIE or MPE
31 which shows they are highly influenced by: the mixture ratios [81], flow velocity

1 [86],[87],[84],[85], focal volume [88],[89],[82] and pulse duration [79], however
2 the present study does not involve measurement of MIE.

3 **1.1.7.3 Ignition studies: Flame kernel investigations**

4 The particular focus of this study is the flame kernel development which is the
5 transition from an ignited spark kernel to a self-sustained flame. Understanding
6 the characteristic behaviour of the flame kernel is of high importance because a
7 successful ignition event relies on the survival of the flame kernel.

8 In one of the first laser ignited flame kernel propagation studies, Santavicca et al.
9 [90], used pulsed laser shadowgraph to visualize the flame kernel development of
10 methane-oxygen flows (at atmospheric pressure) at different equivalence ratios
11 for laminar to turbulent flows. In this study, they measured the flame kernel radius
12 as a function of time and equivalence ratio and compared laser ignition results
13 with that obtained from a General Motors electric ignition system. From their
14 results, they concluded that the laser ignition system performed better than the
15 electric ignition system. In addition, they observed that the measured initial flame
16 kernel Initial radius correlated well with the predicted Energy from Taylor's
17 spherical blast wave model [91]. Please refer to equation 2.15 -2.18 in section
18 2.2.3.2 for details of the spherical blast wave theory.

19 In Spiglanin et al. [92], OH PLIF was used to measure the flame kernel
20 development in LIS ignited hydrogen-air mixtures as a function of gas
21 compositions and time. In this study, a small Q-switched Nd:YAG laser was used to
22 produce 8 ns pulses. They observed that early flame kernel growth is dominated
23 by gas motion induced by the short duration spark. They concluded that the
24 ultimate fate of an ignition event lies with the chemistry of the reactions which
25 determines whether the gas undergoes a transition from hot plasma to
26 propagating flame.

27 Phuoc and White [93] measure plasma dimensions as a function air-fuel ration in
28 laser-ignited methane-air mixture at 1 atm. With a MIE of 3-4 mJ, they observed
29 the spark length and radius were about 0.8 and 0.3 mm, respectively. Similar to
30 other studies, the plasma elongated in the direction of laser beam. The shape of
31 laser spark was oval and lean for rich methane-air mixture whereas it becomes

1 cylindrical in shape for stoichiometric and near stoichiometric methane–air
2 mixture.

3 Bindhu et al. [94] investigated the flame kernel development from a laser-induced
4 spark in argon. They found out that at increasing gas pressures, the plasma can
5 absorb the incident laser energy more effectively. This means that the transmitted
6 energy through the focal volume is less, and the laser ignition and flame kernel
7 propagation is more effective.

8 Lackner et al. [95] characterized flame kernel development in stoichiometric and
9 lean CH₄/Air at 10 bar using Schlieren photography and OH PLIF. From the
10 Schlieren results, a faster evolution was observed from the stoichiometric mixture
11 while OH PLIF image showed the toroidal and front lobe shapes in the early flame
12 kernels.

13 Beduneau and Ikeda [96] investigated the laser-induced spark kernel in a
14 premixed laminar methane–air burner. They observed an asymmetric toroidal
15 shape of the flame kernel, which is caused by the expansion mode of the shock
16 wave. In their explanation, they attributed the asymmetric behaviour to be partly
17 due to the plasma characteristics. In addition, they observed that flame kernel
18 growth initial stages were strongly influenced by spark energy while at the later
19 stages, kernel growth depended mainly on the relative air–fuel ratios. In a later
20 study of basic firing and non-firing phenomena, Beduneau et al., 2009 [97] showed
21 that the level of radical formed inside the flame kernel is a key factor for the
22 sustainability of ignition.

23 Bradley et al. [20] studied ignition of gaseous propane-air pre-mixture in an
24 explosion bomb using high-speed Schlieren photography. They observed that gas-
25 dynamics of the shock waves and high energy plasma leads to an overdrive in the
26 laminar flame speed to such an extent that it decays from elevated values contrary
27 to normal flames. From this observation, they concluded that near the LFL, the gas-
28 dynamic effects can stretch the flame to extinction and narrow the ignition limits.

29 Srivastava et al. [98] employed Shadowgraph technique to visualize the flame
30 kernel shape and propagation of laser-ignited CNG–air mixture inside constant
31 volume combustion chamber, with the aim of simulating end of Compression

1 stroke conditions of a SI engine. They observed the flame kernel shape to be
2 structurally identical for all air–fuel ratios measures. However, they observed a
3 decrease in both flame velocity and peak pressures for lean mixtures and
4 concluded that laser ignition system applied engines are unable to deal with
5 CNG/air mixtures leaner than $\frac{1}{\phi} = 1.6$.

6 In Dharamshi et al. [99], shadowgraph technique was used to visualize flame
7 kernel evolution and the effect of changing air-fuel (H_2) ratio in a CVCC at initial
8 pressure 10 bar and initial temperature 373 K. From the results, they observed
9 that the flame speed decreased with increasing air-fuel ratio in all directions. Also,
10 they observed multiple wave fronts on the toroidal surface for richer mixtures
11 than leaner mixtures. This explains the comparative difference in intensity of
12 explosion in both rich and lean mixtures. In addition, they observed increase in the
13 required MPE for leaner hydrogen-air mixtures and that by increasing the laser
14 pulse energy, time taken to attain peak cylinder pressure reduced slightly,
15 indicating faster flame speeds, however the peak cylinder pressure remained
16 almost constant. Their conclusion was that laser ignition is a potential enabling
17 technology in realizing the dream of a practical hydrogen fuelled engine.

18 Mulla et al. [100] of University College London, showed how the influence of
19 mixture composition, flow velocity and equivalence ratio on the flame-kernel
20 perimeter growth of CH_4 /air using OH-PLIF technique. From this investigation,
21 they observed a distinct shift in the trends of evolution of kernel perimeter near
22 the lean flammability limit in CH_4 /air and H_2 /air mixtures. Also, they observed
23 that the flow velocity (in both laminar and turbulent flow regimes) did not have a
24 significant influence until a certain time was reached. This they attributed to
25 competing effects, between the strain rate and turbulence.

26 In Yu et al. [101] the effect of increasing laser repetition rate (i.e., 1 Hz, 100 Hz and
27 250 Hz) on the stabilization of premixed methane-air flame kernel were
28 investigated using CH^* chemiluminescence. This study is typical example of plasma
29 assisted combustion where high frequency laser pulse can be used to achieve flame
30 stabilization. Initially, they observed the plasma energy coupling under these
31 conditions and discovered it was not affected by the air flow. From measurements

1 of catch-up distance and time of merging of consecutive flame kernels, they
2 observed continuous combustion flame stabilization for repetition rates of 100 Hz
3 and 250 Hz. In addition, they observed that the flame kernel formed by the last LIP
4 does not affect the evolution of the newly formed flame kernel by the next LIP.
5 Their conclusion was that a higher laser repetition rate will lead to a shorter catch-
6 up distance which is beneficial for flame stabilization.

7 In another example of plasma assisted combustion, Dumitrache et al. [48] studied
8 the impact of using a dual frequency pulse on the flame kernel growth of laser
9 ignited lean propane-air mixture. By using OH* chemiluminescence, they
10 compared the flame kernel evolution of a dual-pulse LISI ($\lambda = 266$ nm and $\lambda =$
11 1064 nm) with that of a single-pulse LISI ($\lambda = 1064$ nm). For single-pulse laser-
12 ignited lean mixture, they observed detachment of the third lobe flame kernel due
13 to high strain rates that which may extinguish the flame. For double-pulse case, no
14 detachment was observed due to the existence of a beam waist offset whereby the
15 resulting vorticity suppresses formation of the third lobe, consequently reducing
16 flame stretch. Their conclusion was that the dual-pulse method enables reduced
17 flame speeds (at early times), an extended lean limit, increased combustion
18 efficiency, and decreased laser energy requirements.

19 **1.1.7.4 Research gaps and challenges**

20 The essence of most experimental studies on ignition is to provide validation data
21 for the development of mathematical ignition models which can be extended to
22 simulate real combustion situations. To have accurate models of laser ignition,
23 data on the physical characteristics of the flame kernel must be available for
24 different fuel/air mixtures as well as their parametric dependencies. However,
25 following the extensive literature survey, a number of research gaps and
26 challenges still persist. Below are some of the challenges and gaps identified which
27 this study hopes to address.

- 28 • Most laser ignition measurements observed in the surveyed literatures were
29 conducted in either open atmospheric burners [96], [92], [85] or closed
30 combustion vessels [93], [20], [98]. The advantage of combustion vessels is that
31 a quiescent mixture can easily be obtained, however their designs are more

- 1 complex and providing optical access require very expensive materials. Such
2 complexities are eliminated with the use of atmospheric burners which justifies
3 why it is chosen in the present work.
- 4 • A significant number of the reviewed literatures were focussed on performance
5 of the combustion systems whereas knowledge of the flame kernel structure is
6 still limited. This raises the question of what the key features of a developing
7 flame kernel are and how useful are they in predicting ignition success or
8 failure. Although the features of the flame kernel were observed by past
9 researchers [92], [20] through photographic imaging, there is no consensus as
10 to the origin of these features and their link to ignition success or failure which
11 necessitates further investigation of these salient features using robust optical
12 techniques.
 - 13 • While photographic images may provide proof of an on-going reaction, the
14 observed features may not be enough evidence of a successful ignition since the
15 early kernel growth is mainly influenced by the gas dynamics of the shock wave
16 and the high energy plasma. A necessary feature of an igniting kernel is the
17 production of sufficient quantity of radicals during initiation of chemical
18 reaction. Due to the optical technique employed in most laser ignition studies,
19 only information relating to the fluid dynamics of the flame kernel were
20 provided. To understand the complex processes involved in laser ignition, both
21 the fluid dynamics and chemical kinetics data are required. Therefore, another
22 question which is yet to be answered is what is the coupling between the fluid
23 dynamics of the flame kernel and the chemical reactions occurring?
 - 24 • Another limitation resulting from optical the optical techniques commonly
25 employed is that most techniques only provide information about the physical
26 characteristics of the kernel with no evidence of how the flame is interacting
27 with the flow-field. Such information will be useful for optimising combustion
28 based on ignition location.
 - 29 • Any investigation of the flame kernel characteristics that excludes the effects of
30 operational parameters like pulse energy and flow velocity would be
31 incomplete. The laser pulse energy is a key parameter that could affect the

1 efficiency of the flame kernel formation and its subsequent propagation to a
2 stabilized flame. Although the effects of laser energy on flame velocity has been
3 studied previously [102],[99], its effect on the early flame kernel development
4 has not been properly addressed. In most combustor designs, variability of the
5 combustion process is a continuing problem which arises from high turbulence
6 and high turbulence is associated with high flow velocity. Hence, the need to
7 address the question of the extent of flow velocity influence on the
8 development and propagation of the flame kernel.

- 9 • In terms of the fuels investigated till date, there has been more focus on
10 methane [88], [103],[104],[105] which provides a good baseline for gaseous
11 fuels due to its unique properties. There has been some investigations of other
12 fuels like hydrogen [99], propane [20], etc, however this is only a small number
13 considering the large number of available gaseous fuels. Since each fuel differs
14 from the rest in their thermochemical properties, there is also the need to
15 study what impact the fuel properties have on the development characteristics
16 of the laser flame kernel.

17 **1.1.8 Thesis Objectives and Contributions**

18 The current PhD work involves detailed investigation of the development
19 characteristics of a LISI flame kernel in flowing premixed fuel/air mixtures. The
20 goal is to show the possible link between these characteristics and ignition success
21 as well as possible use of these data for development and validation of laser
22 ignition models. The investigation would be explored under three specific
23 objectives, each with the aim of addressing specific questions raised from gaps
24 identified in literature.

- 25 • The first objective is to investigate the flow field characteristics of a developing
26 LISI flame kernel in both laminar and turbulent flow regimes. To accomplish
27 this objective methane mixed with Mie-scattered air forming a stoichiometric
28 aerosol mixture is flowed through a co-axial straight tube burner and then
29 ignited by a single pulse laser beam of 1064 nm wavelength and 7 ns pulse
30 width. To enable visualization of the reacting flow field, a combination of Laser

1 sheet tomography and High-Speed Particle Image Velocimetry (HSPIV)
2 techniques was employed. This allowed simultaneous measurement of the
3 flame kernel geometry and the flow velocity with the hope that this would
4 address the problem of shortage of data on how the flow-field is interacting
5 with the developing flame kernel. By observing the flow field features before
6 and during the ignition event, combustion can be optimized based on ignition
7 location which is one of the advantages of laser ignition over the spark plug
8 [69].

- 9 • The second objective is to investigate the characteristics of the LISI flame
10 kernel based on the mean OH* chemiluminescence. To accomplish this
11 objective stoichiometric mixture of CH₄/Air flowing through a burner with co-
12 axial contracting nozzles were repeatedly ignited by focused laser beam (1064
13 nm wavelength and 2 Hz frequency). Through direct capture of the kernel OH*
14 chemiluminescence with CCD camera at different time delays (0.05 – 10 ms),
15 the geometric data which is linked to the fluid dynamics of kernel would be
16 extracted. In addition, the luminosity of the OH* which is linked to the chemical
17 kinetics would be obtained. Both data could be used to address the problem of
18 shortage of data on the coupling between the fluid dynamics and chemical
19 kinetics of the laser flame kernel. The study scope will also include the effect of
20 changing pulse energy and flow velocity with the aim of addressing what
21 influence these operational parameters have on the flame kernel development.
- 22 • The third and final objective is to investigate the effect of changing fuel
23 properties on the development characteristics of the flame kernel. The study is
24 divided in three sub-sections. First, the study will characterise the flame kernel
25 based on differences in the equivalence ratios of the fuel which by implication
26 means differences in both the adiabatic flame temperature and laminar flame
27 velocity of the fuel. Next, the sensitivity of the flame kernel characteristics to
28 the fuel composition is investigated for three different fuels (i.e., methane,
29 propane, ethylene) at constant adiabatic flame temperature. Lastly, the
30 sensitivity of the flame kernel characteristics to the fuel composition is
31 investigated for the three fuels at constant laminar flame velocity. The

1 experimental setup and diagnostic technique employed will be the same as that
2 used in achieving the second objective.

3

4 **1.1.9 Thesis Outline**

5 The entire thesis consists of eight chapters beginning with the current
6 introductory chapter.

- 7 • Chapter 2 presents a review of the background theories which form the basis
8 for understanding Laser ignition and its propagation in gaseous fuel/air
9 mixtures. Most of the important flow properties as well as the processes
10 involved in laser induced spark ignition are covered.
- 11 • Chapter 3 presents details of the experimental setups and the diagnostic
12 techniques employed in this work. First, a description of the burners and flow
13 system configurations used for the laser ignition study were given. In addition,
14 a description of the optical systems and processing steps used for
15 implementing Laser tomography, PIV and chemiluminescence techniques were
16 also provided.
- 17 • 3.6 presents a study of the characteristics of the flame kernel development
18 within its flow field. Results of the kernel geometry and its flow field features
19 obtained by 2D Laser tomography are presented. In addition, the flow field
20 structures obtained using PIV are also reported.
- 21 • Chapter 5 presents a study of the flame kernel characteristics based on the
22 OH* chemiluminescence. The results include the visual images of the flame
23 kernel as well as quantitative analysis of the flame front propagation and OH*
24 luminosity. This investigation was extended to include effects of laser energy
25 and flow velocity as described in the objectives.
- 26 • In Chapter 6 the investigation was extended to include the effects of varying
27 mixtures ratios and fuel types on the chemiluminescence characteristics of the
28 flame kernel.

- 1 • Chapter 7 presents a discussion of all the results from the investigation in a
2 coherent and insightful manner.
- 3 • Chapter 8 presents the conclusion of the findings from the investigations. A
4 recommendation of possible future requirements to better understand the
5 research is also given.
- 6 • The report also includes References and four Appendices. Appendix A:
7 Experimental Procedures provides further information on the experimental
8 procedures adopted while conducting the experiments. Appendix B: Risk
9 Assessment is the risk assessment document developed for the experimental
10 campaign. Appendix C: MATLAB Codes Used to Analyse Laser Tomography
11 Images and PIV Data contains the code developed for analysis of the tomographic
12 and PIV data. Appendix D: MATLAB Code Used to Analyse
13 Chemiluminescence Data contains the code developed for analysis of the
14 chemiluminescence data. Further sensitivity analysis of the kernel characteristic
15 parameters to different test conditions are provided in Appendix E: Sensitivity
16 Analysis.

1 Chapter 2 Flow and Laser Ignition Fundamentals

2 This chapter provides the theoretical basis for understanding Laser ignition and its
3 propagation in gaseous premixed fuel-air mixtures. It begins with a discussion of
4 the important properties of the flow in both reacting and non-reacting state. Then
5 the fundamental theories of laser ignition and flame propagation are presented.

6 2.1 Fundamental Properties of Non-reacting and Reacting Gas Flows

7 Characterization of the flow turbulence structures is very important since they
8 influence both the development and continued burning of the flame. Before any
9 attempt is made to investigate the influence of the gas flow mixture on the
10 propagation of the flame kernel, it is important to understand the fundamental
11 properties of the flow mixture in both reacting and non-reacting state.

12 2.1.1 Non-reacting Flow properties

13 Generally non-reacting gas flows are classified into *laminar* and *turbulent* flows.
14 Laminar flow is an ordered motion of the fluid particles along a streamline (or
15 approximately parallel flow contour lines) which occurs when the velocity of flow
16 is sufficiently reduced for a fixed diameter nozzle. In contrast turbulent flow which
17 is the more practical case is characterized by random or irregular motion of fluid
18 particles due to high velocity. In such a flow, the rates of fluid transfer and mixing
19 are several times greater than the rates of molecular diffusion. To distinguish
20 between the two flow types, *Reynolds Number (Re)* is used. The Reynolds Number
21 is defined as the ratio of the inertia force to the viscous force acting on an element
22 of fluid [106]. It relates to the characteristic length (L), mean velocity (U), fluid
23 density (ρ) and dynamic viscosity (μ) in a dimensionless form given by equation
24 2.1. It is widely accepted that the flow is laminar if $Re < 2000$ and turbulent if
25 $Re > 4000$.

$$1 \quad Re = \frac{\rho UL}{\mu} \quad 2.1$$

2 In characterizing turbulence flows, statistical properties are used because the flow
3 is usually viewed as random local fluctuating motions superimposed on a uniform
4 or mean motion of the fluid. The main properties of fluid motion are *instantaneous*
5 *and mean velocity, fluctuating velocity or turbulent intensity, turbulent length*
6 *scales and turbulent time scales.*

7 **Instantaneous velocity:** In a steady turbulent flow situation, the
8 instantaneous local fluid velocity, u (in a specific direction) is given as the
9 sum of the mean velocity component, \bar{u} and the fluctuating velocity
10 component, u' (also known as turbulent intensity).

$$11 \quad u = \bar{u} + u' \quad 2.2$$

12 **Mean velocity:** this is the time average value of the instantaneous velocities
13 obtained in a steady flow situation. In quasi-periodic flows such as
14 obtainable in engine cylinders, the mean flow varies from cycle-to-cycle,
15 hence measurements made over many cycles are *phase-averaged* to obtain
16 the ensemble-averaged velocity [41].

$$17 \quad \bar{u} = \frac{1}{\tau} \sum_{t_0}^{t_0+\tau} u(t) \quad 2.3$$

18 **Turbulence intensity:** this provides a measure of the turbulence level
19 present in a flow field. It is the root-mean-square, RMS value of the velocity
20 fluctuations, u' divided by the mean velocity of the fluid motion, \bar{u} [41].

$$21 \quad u' = \sqrt{\sum (u - \bar{u})^2} \quad 2.4$$

22 **Turbulence length scales:** this provides a measure of the size of turbulent
23 eddy structures present in the flow field. The length scales of turbulence
24 include Integral length scale (i.e., largest size eddies), Taylor micro-scale
25 (i.e., medium size eddies) and Kolmogorov length scale (i.e., smallest size
26 eddies). The Integral length scale (I) and Kolmogorov length scale (η) can
27 be approximated as below [107].

1
$$I = \frac{u'^3}{\varepsilon} \quad 2.5$$

2
$$\eta = \left(\frac{u'^3}{\varepsilon}\right)^{\frac{1}{4}} \quad 2.6$$

3 **Turbulence time scales:** this provides a measure of the time over which the
4 turbulent structures occur. An example is the Integral time scale (τ) given
5 by [107]:

6
$$\tau = \frac{\text{integral scale } (l)}{\text{turbulence intensity } (u')} = \frac{u'^2}{\varepsilon} \quad 2.7$$

7 2.1.2 Reacting Flow properties

8 Gas mixtures consisting of fuel and oxidant undergo unsteady reactions when
9 ignited. Ignition is defined as a time-dependent process in which a starting
10 reaction of fuel-oxidant mixture evolves until a steadily burning flame is reached
11 or the mixture has completely reacted [27]. In internal combustion engines, this
12 process can occur in either of two ways: autoignition or spark-induced ignition. In
13 autoignition the mixture is compressed so that its temperature rises above the
14 autoignition temperature where decomposition and radical formation occurs to
15 such an extent that chain branching can start and proceed at a significant rate[27].
16 In contrast, spark induced ignition involves the generation of a plasma within a
17 small fractional volume of the mixture that yields the initial radicals via ionic
18 processes. At this early ignition stage, the shape of the flame front is called the
19 flame kernel, and it can be described as the transition stage from the plasma to a
20 self-sustaining flame[27].

21 Depending on the state of the mixture prior to ignition, two distinct forms of
22 ignition reaction occur. The first is premixed charge ignition which occurs when
23 the fuel and oxidant are completely mixed before ignition occurs. The second case
24 in which there is no prior mixing of fuel and oxidant is known as non-premixed
25 charge ignition. This is the ignition type which this work will be focussed on. Below

1 are some of the fundamental properties of a premixed flow mixture which affects
2 its propagation:

3 **Equivalence ratio (ϕ):** is a fundamental property of the flow mixture that
4 characterizes the composition of fuel and air. It is defined as the given fuel/air
5 mixture ratio divided by the corresponding fuel/air mixture ratio at stoichiometric
6 composition [27].

$$7 \quad \phi = \frac{x_f/x_a}{(x_f/x_a)_{stoic}} \quad 2.8$$

8 where: x_f is the mole fraction of the fuel, x_a is the mole fraction of the air. Lean
9 mixture has values of ϕ less than one while rich mixtures have values of ϕ greater
10 than one. A mixture of stoichiometric composition is defined by a value of ϕ equal
11 to one.

12 **Flammability limits:** define the range of equivalence ratio of fuel/air mixture at
13 specified temperature and pressure within which an ignited flame kernel will
14 continue to propagate even after the ignition source has been extinguished. The
15 maximum value in the rich mixture range is known as the upper flammability limit
16 (UFL) while the minimum value in the lean mixture range is known as the lower
17 flammability limit (LFL). The flammability maps of fuels are usually determined
18 experimentally, and they depend on both temperature and pressure. Table 2.1
19 shows the results of flammability limits of the investigated fuels, measured in an
20 explosive vessel with temperature and pressure conditions maintained at 35°C and
21 5 psi, respectively. A typical fuel flammability map and its dependence on
22 temperature is shown in Figure 2.1. According to the figure, the flammability range
23 for a mixture increases with temperature and finally reaches a temperature
24 beyond which autoignition will occur. At higher pressures, the flammability
25 window will usually exceed the limits shown [27]. These limits are also affected by
26 the nature of the fuel, direction of flame propagation, size and shape of combustion
27 chamber, temperature and pressure [108].

28 **Adiabatic flame temperature (T_{ad}):** this is the temperature achieved when a
29 combustible mixture of fuel and air burns to completion under isobaric and

1 adiabatic condition. In other words, it is the maximum temperature of the flame in
 2 the absence of non-unity Lewis number, differential diffusion and strain
 3 effects[109]. According to [109], the adiabatic flame temperature through the
 4 Arrhenius kinetics exerts a dominant influence on flame speed. Theoretically, T_{ad}
 5 is obtained from the overall energy balance based on the one-step Arrhenius
 6 kinetics:

$$7 \quad T_{ad} = T_u + \frac{QY_u}{c_p} \quad 2.9$$

8 where: T_u is the unburned gas temperature, Y_u is the mass fraction of the deficient
 9 reactant, Q is the heat released per unit mass fraction of the deficient reactant, c_p
 10 is the specific heat of the mixture at constant pressure.

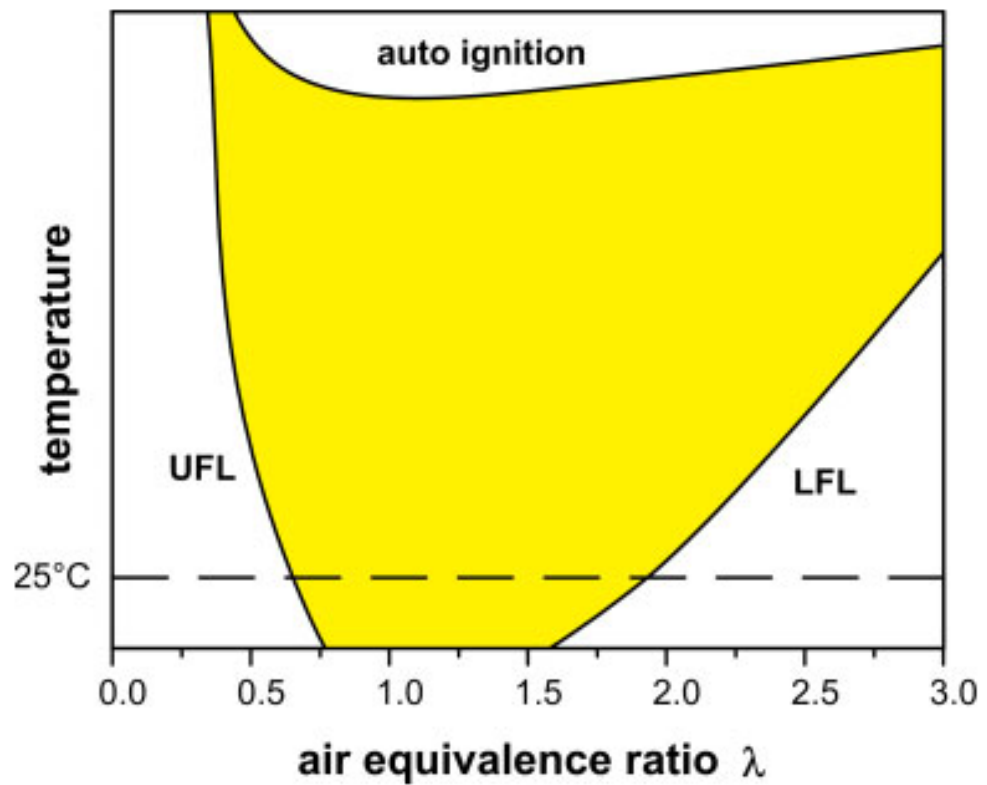
11 Figure 2.1 shows a comparative T_{ad} versus ϕ values obtained using GASEQ
 12 chemical equilibrium software [110] for the different fuel gases studied in the
 13 present work. As shown in the figure, T_{ad} peaks at slightly above $\phi = 1$ for all the
 14 fuels. Of the three fuels, ethylene has the highest flame temperature for a given ϕ
 15 value while methane has the lowest. The difference in values is due to the different
 16 enthalpy of reaction required to dissociate the bonds. For example, the respective
 17 bond strength of methane and ethylene molecules are 436 and 699 kJ/mol.

18 **Laminar burning velocity (S_L):** this is the speed at which a planar flame travels
 19 into the mixture in the absence of heat losses. It is also defined as the velocity of
 20 the combustion wave normal to itself and relative to the unburnt gas [52]. It is a
 21 fundamental parameter because it is the only flame speed unique for a gas of a
 22 fixed composition, initial temperature and pressure, without further specification
 23 of hydrodynamic conditions such as stretch rate, Reynolds number, etc [111].
 24 Hence, accurate determination of S_L is of fundamental importance since it is
 25 required for the development and validation of kinetic mechanisms for fuels.
 26 Experimental measurement of S_L is based on four main techniques that have
 27 proven to be quite versatile and accurate. These includes Bunsen flame approach,
 28 Flat flame approach, spherically expanding flame method and Stagnation flame
 29 method. Figure 2.2 compares S_L versus ϕ measurements obtained for the three

- 1 fuels by using the Stagnation flame method [112]. In addition, S_L can be derived
- 2 theoretically using the chemical structure of the flame.
- 3 **Table 2.1: Flammability limits observed from experiment [113].**

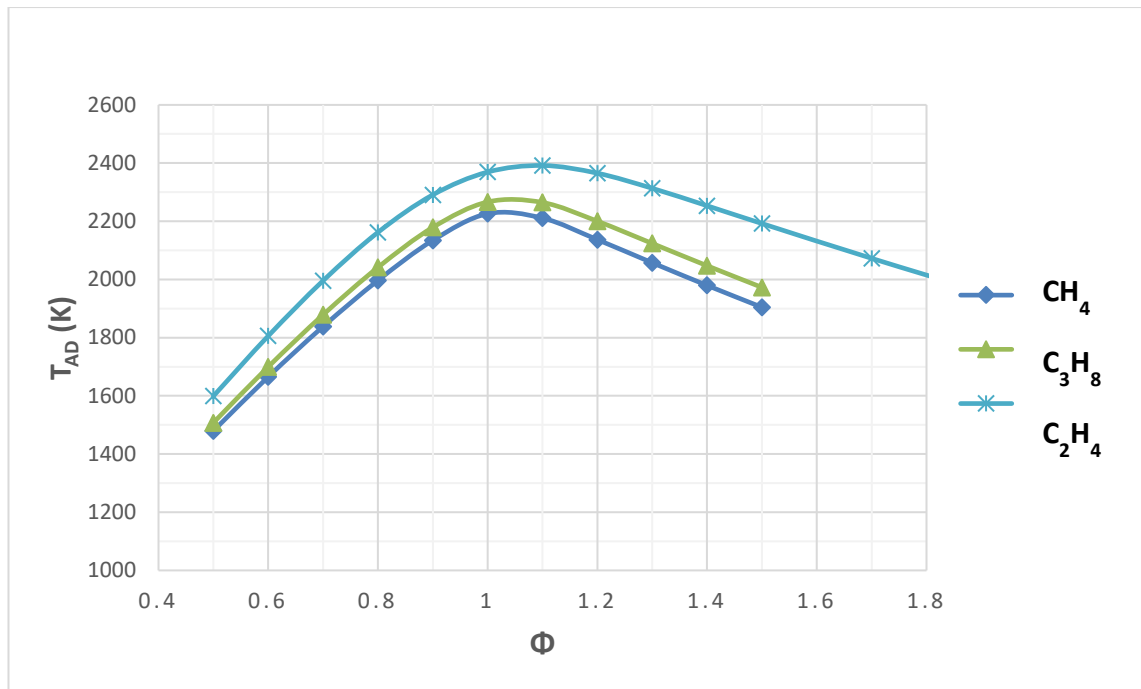
Property	Methane	Propane	Ethylene
LFL - UFL (%vol)	4.9 - 15.8	2.03 - 10.0	2.74 - 31.5
LFL - UFL (ϕ)	0.50 - 2.00	0.50 - 2.64	0.40 - 6.57

4

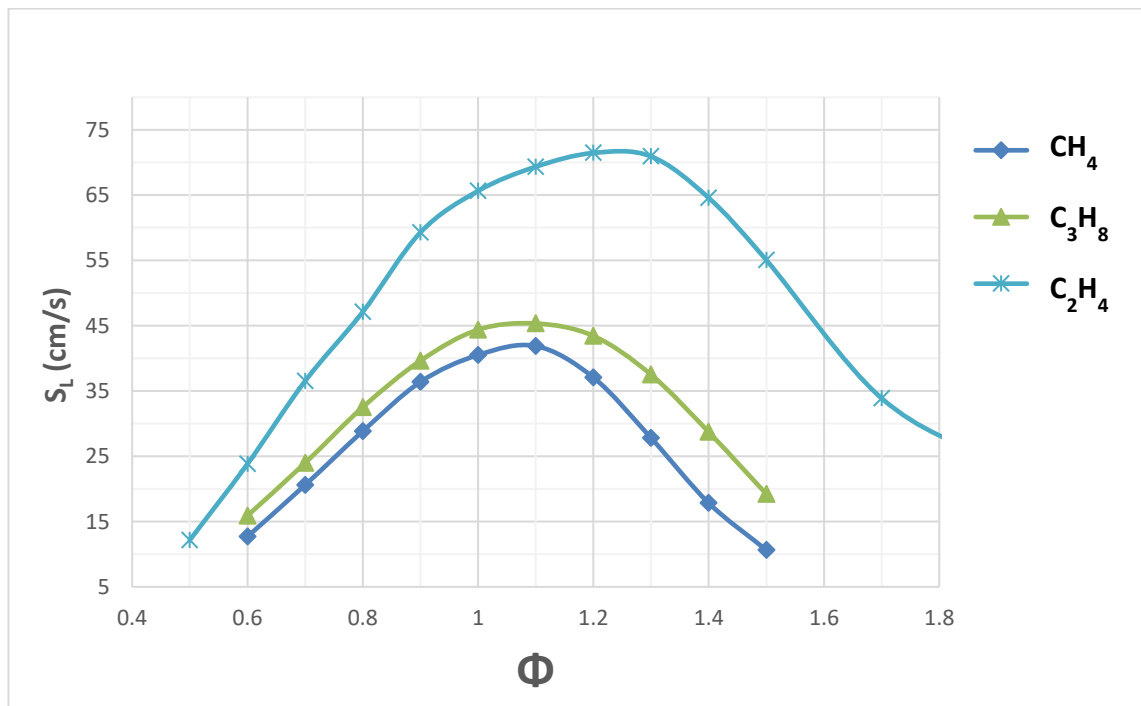


5

- 6 **Figure 2.1: Typical flammability map showing its dependence on temperature and**
- 7 **the autoignition limits[27].**

1
23 **Figure 2.2: Adiabatic flame temp versus Equivalence Ratios for CH_4 , C_3H_8 and C_2H_4**
4 **(Source: [110]).**

5



6

7 **Figure 2.3: Lam flame velocity versus Equivalence Ratios for CH_4 , C_3H_8 and C_2H_4**
8 **(Data Source: [112]).**

9

1 **2.2 Fundamentals of Laser Induced Spark Ignition**

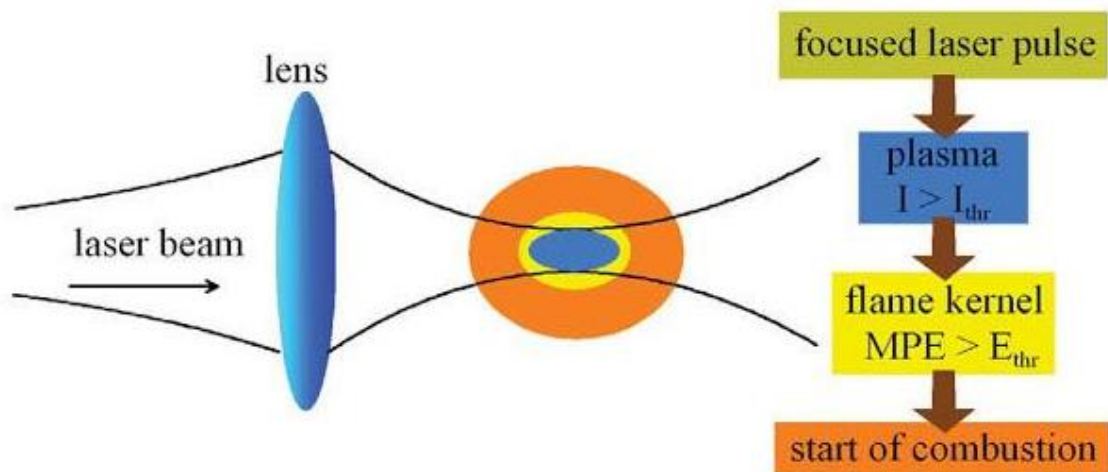
2 **2.2.1 Mechanisms of Laser Ignition**

3 According to Ronney [10], there are four known mechanisms by which laser
4 radiation can ignite combustible gas mixtures. These includes thermal initiation,
5 non-resonant breakdown, resonant breakdown, and photochemical ignition. In
6 thermal initiation, no electrical breakdown is required, instead a laser source is
7 used to heat up the mixture beyond its threshold ignition temperature[16]. It is
8 also possible by heating of a target surface in the combustion chamber.
9 Consequently, molecular bonds are broken, and chemical reaction occurs leading
10 to ignition. This mechanism is unique in that it can easily be used to ignite
11 combustibles in combination of solid [114], [115], liquid [116], and gas phases
12 [117], [118],[119],[120]. The method is most suitable for fuel/oxidizer mixtures
13 with strong absorption at the laser wavelength, but it involves long ignition delay
14 times. In photochemical ignition, laser photons dissociate target molecules into
15 highly reactive radical species, leading to ignition (provided radicals production
16 rate is higher than their recombination rates). Various studies on this ignition
17 mechanism have shown that it involves less energy and it does not involve
18 photoionization [121],[122],[123],[124] or direct heating [125]. Compared with
19 other mechanisms, mixtures can easily be ignited at lower pressures and closer to
20 the flammability limits. However, it requires a close match between the laser
21 excitation wavelength and the target molecules absorption wavelength; hence a
22 tuneable laser is required. The resonant breakdown ignition process involves, first,
23 a non-resonant multiphoton dissociation of molecules resulting to freed atoms,
24 followed by a resonant photoionization of these atoms. This process generates
25 sufficient electrons needed for gas breakdown. Theoretically, less input energy is
26 required due to the resonant nature of this method. This mechanism has been
27 demonstrated for successive ignition of H_2/O_2 and H_2/N_2O mixtures at
28 atmospheric pressure using a tuneable UV laser to produce laser pulses near 225.6
29 nm wavelength and to produce ground state oxygen atoms from the two-photon
30 photodissociation of O_2 and NO_2 molecules [18],[19],[126]. The Non-resonant
31 breakdown ignition is the most widely studied laser ignition method with similar

1 characteristics to the electric Spark Ignition [127]. It involved the use of focused
 2 laser beam to create an electric field of sufficient intensity which cause dielectric
 3 breakdown of the air-fuel mixture. Like the electric spark system, it involves
 4 production of plasma which emits light, heat and shockwave. Hence, it is also
 5 referred to as Laser Induced Spark Ignition (LISI) or simply Laser Ignition (LI). In
 6 the rest of the thesis, the terms Laser Ignition or Laser Induced Spark Ignition
 7 would be used to refer to Non-Resonant Breakdown ignition.

8 2.2.2 Basic setup and important thresholds for successful laser ignition

9 The basic setup needed for laser ignition is shown in figure 2.3. It consists of a
 10 short laser pulse beam (usually in the *ns* or *ps* time regime) which is tightly
 11 focused within a small volume of a flammable mixture by a converging lens.



12

13 **Figure 2.4: Schematic of Laser ignition setup and thresholds for ignition success**
 14 [11]

15

16 Also depicted in figure 2.3 are the important thresholds for successful laser
 17 ignition most of which are discussed in the paragraphs below.

18 **Breakdown Intensity threshold (I_o):** The first condition for a successful ignition by
 19 laser induced breakdown is that in the focal point, a certain intensity threshold (I_o)

1 has to be exceeded. If the laser pulse exceeds this intensity, a plasma is formed, and
 2 it is possible to ignite a stoichiometric mixture with pulse energies below 1 mJ. For
 3 most gases under atmospheric condition, the required threshold for plasma
 4 generation is of the order of 10^{10} - 10^{12} W/cm² [27], [35].

5 **Minimum Ignition Energy (MIE or E_{min}):** A second necessary condition for
 6 successful ignition is that the energy input from the laser spark must exceed a
 7 certain value known as minimum ignition energy. This is the energy required to
 8 raise the temperature of a sphere of gas with radius equal to the characteristic
 9 flame thickness (δ_f) and temperature equal to the adiabatic flame temperature
 10 (T_{ad}) [10],[27]. For deflagrative combustion, the MIE and δ_f can be approximated
 11 by the following equations:

$$12 \quad E_{min} = \frac{4\pi\delta_f^3}{3} * \rho_f * c_p * (T_f - T_g) \quad 2.10$$

$$13 \quad \delta_f = \frac{k}{\rho_f * c_p * v_f} \quad 2.11$$

14 where: ρ_f, T_f, v_f represents the density, temperature, and velocity of the flame,
 15 respectively. T_g, c_p represents the cold mixture temperature and specific heat
 16 capacity at constant pressure.

17 The concept of MIE had earlier being captured in [128] description of ignition of
 18 premixed gases by deflagrations (i.e., flame propagation at sub-sonic speed) as
 19 follows:

20 "If a subcritical quantity of energy (in the form of heat and/or radicals) is
 21 deposited in a combustible mixture (of premixed gas and air), the resulting
 22 flame kernel decays rapidly because heat and radicals are conducted away
 23 from the surface of the kernel and dissociated away from the surface of the
 24 kernel and dissociated species recombine faster than they are regenerated
 25 by chemical reaction in the volume of the kernel. The kernel extinguishes
 26 after consuming a small quantity of reactant. However, if the ignition energy
 27 exceeds a certain threshold (i.e., the minimum ignition energy) at the time
 28 when the peak temperature decays to the adiabatic flame temperature, the
 29 temperature gradient in the kernel is sufficiently shallow that heat is

1 generated in the kernel faster than it is lost due to conduction to the
2 unburned mixture”.

3 The problem with the above description (also known as Homogeneous hot-gas
4 model) is that it excludes gas breakdown and blast wave processes present in laser
5 ignition. Although, the description by Lewis and von Elbe was originally applied to
6 electrical sparks, it is equally applicable to laser-ignited combustible gas mixtures
7 and has been measured by many authors.

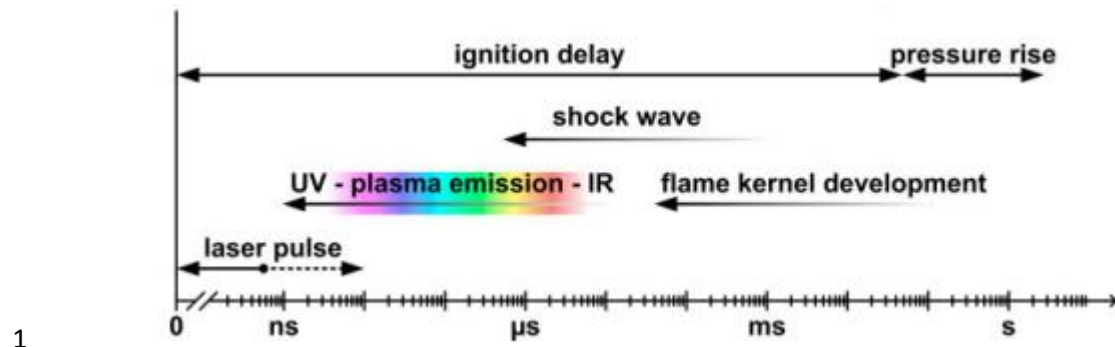
8 **Minimum Pulse Energy (MPE):** this is the minimum value of total pulse energy
9 needed to yield ignition. The MPE includes both the MIE and the losses due shock
10 waves, reflections, convections and radiations. It is a more practical term
11 compared with MIE which is prone to experimental errors and uncertainties. For a
12 nanosecond laser pulse, the MPE for methane/air is about 5-10 mJ under lean
13 conditions [11].

14 **2.2.3 The Laser Ignition process**

15 Figure 2.4 gives a picture of the processes involved in a typical Spark Ignition
16 engine and their timescales. As shown in the figure, the processes leading to the
17 formation of a laser-ignited flame can be divided into these four stages:

- 18 • Gas breakdown or Spark generation
- 19 • Spark evolution
- 20 • Flame-kernel development
- 21 • Combustion.

22 While this study is mainly focussed on understanding flame kernel development,
23 efforts will be made in describing some of the other processes. Further details of
24 the processes leading to LIS ignition can be found in [69], [20], [27].



1

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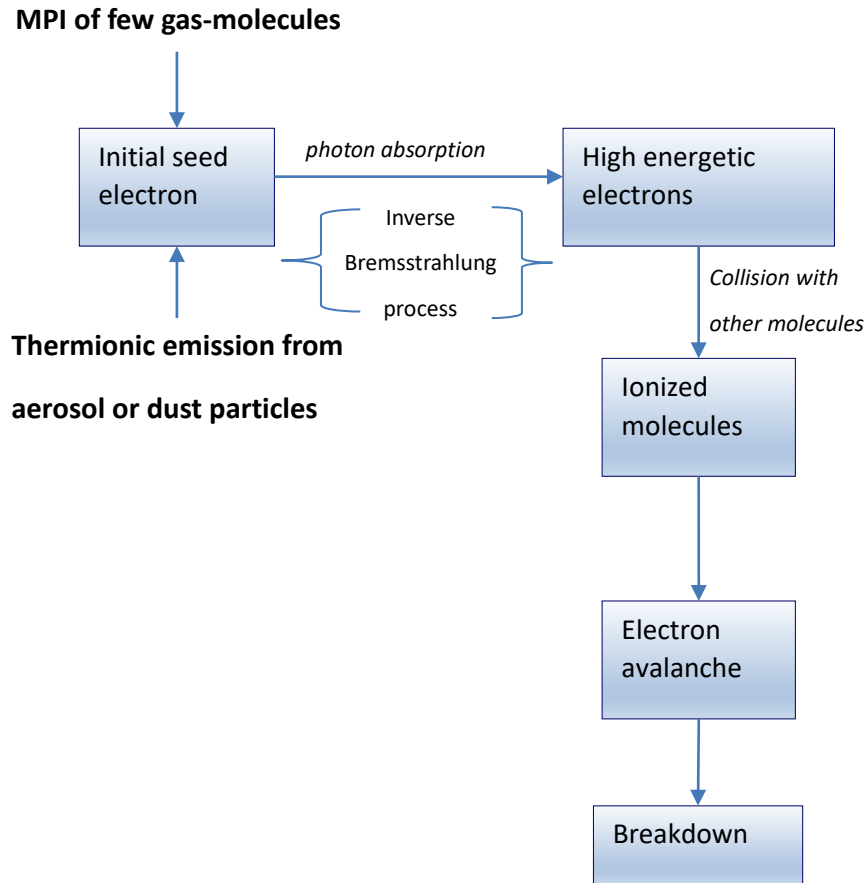
Figure 2.5: Timeline and Sequence of Laser Ignition process [27]

3

4 2.2.3.1 Gas Breakdown (Spark generation process)

5 The first significant event in Laser ignition is the generation of spark plasma by a
 6 tightly focused laser beam. Plasma formation is the result of gas breakdown, and
 7 this happens when the intensity of radiation within the focal volume exceeds the
 8 Breakdown Intensity threshold (I_0) of the gas present. Figure 2.6 shows the steps
 9 involved in laser induced breakdown of gases starting from the initial seed
 10 electrons to the gas breakdown. As shown in the figure, the presence of initial
 11 electrons in the focal volume is a necessary condition for the electron avalanche
 12 (i.e., electron density growth) which leads to gas breakdown. The free electron can
 13 be supplied either by seed electrons present in the gas as impurities or by multi-
 14 photon ionization of gas molecules.

15



1
2

Figure 2.6: The Gas Breakdown process [11].

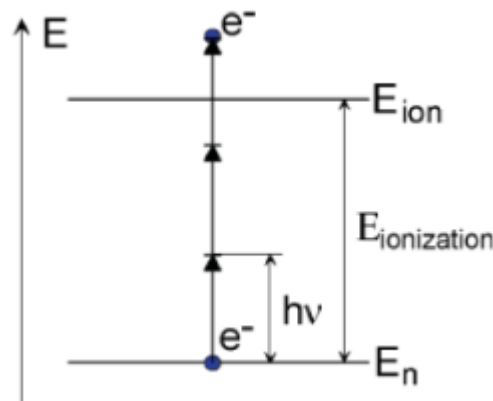
3 **Initial Seed Electron:** Impurities, like aerosol particles, dust or vapor in the focus
 4 can be the source of some initial electrons. Such particles typically have a low
 5 ionization potential (less than 1eV) and, therefore, can be ionized resonantly by
 6 the laser radiation or thermionically due to absorptive heating. Experiments
 7 conducted in air at a wavelength of 10.6 μm , showed that the plasma was initiated
 8 by aerosols within the focal volume[129]. Under normal atmospheric conditions,
 9 there are more than 10^7 particles per mm^3 larger than 0.1 μm [130]. These particles
 10 would heat under laser irradiation by absorption and could generate electrons by
 11 thermionic emission[131].

12 **Multiphoton Ionization:** Another mechanism describing electron generation and
 13 growth is multiphoton ionization (MPI) which is illustrated in Figure 2.7. In the
 14 MPI process, a gas molecule or atom (M) simultaneously absorbs many photons. If
 15 the combined absorbed energy is higher than its ionization potential, then the gas
 16 molecule is ionized (M^+). The reaction process is according to the equation [27]:

$$1 \quad M + m \cdot h\nu = M^+ + e^- \quad 2.12$$

2 where: $h\nu$ = photon energy, m = number of photons necessary to ionize an atom.
 3 For Ionization Energy (IE), m is given as: $m \geq \frac{IE}{h\nu} + 1$. This process is important
 4 only at very short wavelengths (i.e., below $1 \mu\text{m}$) or at very low pressures (i.e.,
 5 below 10 torr), where collisional effects are negligible[132],[35]. It becomes
 6 insignificant at visible and near-IR wavelengths because the photon energy at
 7 these wavelengths is much smaller than the ionization potentials of most gases.
 8 For example, the photon energy for a CO_2 laser is 0.1 eV, and for an Nd-YAG laser at
 9 $1.064 \mu\text{m}$ it is 1.0 eV, while the ionization potentials for most gases are larger than
 10 7 eV. Thus, the multiphoton ionization process would require the absorption of 70
 11 CO_2 photons (or 7 Nd-YAG photons) to ionize most gases.

12



13

14 **Figure 2.7: Illustration of Multiphoton Ionization**[11]

15 **Electron cascade process:** After a sufficient number of free electrons have been
 16 produced via either (or both) of the above processes, the electron cascade process
 17 begins. The electrons gain energy by the absorption of photons which causes them
 18 to accelerate. This process is called inverse bremsstrahlung effect [131]. The
 19 accelerated electrons on impact with neutral gas molecules causes them to ionize
 20 leading to an electron cascade. The reaction process is given as[27]:



1 The resulting effect is known as cascade breakdown, where the number of
 2 electrons (N_e) increases exponentially with time (t) according to the following
 3 equation[27]:

$$4 \quad N_e(t) = N_{e,0} * e^{\left(\frac{t}{\tau_e}\right)} \quad 2.14$$

5 where: $N_{e,0}$ starting electron density.

6 τ_e time constant given as $\tau_e = \frac{1}{\left[v_d\left(\frac{I}{I_0}-1\right)\right]}$

7 v_d diffusion rate of electrons

8 I, I_0 radiation intensity and intensity threshold of medium

9 2.2.3.2 Spark Evolution Leading to Ignition

10 At about 200 ns after the breakdown, the generated spark is completely ionized,
 11 containing highly reactive chemical species that provide energy and are a chemical
 12 source for combustion initiation. At this time, the plasma has an instantaneous
 13 temperature of about 10^5 K and pressure of about 10^3 atm. These extreme
 14 conditions relative to the surrounding ambient gas give rise to rapid expansion
 15 and dissipation of deposited energy. It is also associated with shock wave
 16 generation, which give rise to heat losses in the spark. In the absence of
 17 dissociation, the plasma expansion can be modelled using Taylor's spherical blast
 18 wave theory [133], [91] given by the following equations:

$$19 \quad r = (E_k/\rho_0)^{\frac{1}{5}}t^{\frac{1}{5}} \quad 2.15$$

$$20 \quad v = \frac{2}{5}(E_k/\rho_0)^{\frac{1}{2}}r^{-\frac{3}{2}} \quad 2.16$$

$$21 \quad P = [2/(\gamma + 1)]\rho_0v^2 \quad 2.17$$

$$22 \quad T = T_0 \left[\left(1 - \frac{v_0^2}{v^2}\right) \left(\frac{\gamma-1}{\gamma+1}\right) + 1 \right] \times \left[\left(\frac{v^2}{v_0^2} - 1\right) \left(\frac{\gamma-1}{\gamma+1}\right) + 1 \right] \quad 2.18$$

23 where: r is distance, E_k is the thermal energy behind the blast front generated by
 24 the spark, ρ_0 is the gas density, v and v_0 are the shock and sonic velocities,
 25 respectively, $\gamma = c_p/c_v$, and P and T are the pressure and temperature at the blast

1 front. An illustrative model by [92] for a 10 mJ spark deposited in a stoichiometric
2 H₂/O₂/Ar mixture showed that the spark expansion speed decays from supersonic
3 to subsonic in less than 5 μ s.

4 Based on several studies [92],[20],[80], two scenarios have been proposed to be
5 responsible for transition spark evolution to an igniting flame kernel. The first
6 proposal known as the blast wave ignition model states that ignition is a direct
7 outcome of heat of the surrounding mixture to the ignition temperature by the
8 force of the decaying shock wave. However, available experimental data do not
9 support this model [134],[135]. One reason given is that there is a distinct
10 separation of time scales between the shock wave (several microseconds) and the
11 combustion wave (several milliseconds). Another reason based on Semenov's
12 condition for spontaneous ignition shows that even when there is a shock
13 sufficiently strong to ignite the surrounding combustible mixture, the ignition zone
14 does not remain long enough. The second proposal known as hot gas ignition
15 model states that ignition of the surrounding gas mixture indirectly results from
16 the remaining energy of the hot gas after shock wave expansion. This model is
17 supported by [136] who modelled the ignition coupling by adding an adiabatic
18 expansion model to the blast wave theory. Also, this model did not consider energy
19 losses due to shock wave generation. This is also supported by [92], who modelled
20 the coupling between the spark expansion and ignition by adding a simple kinetic
21 model (which considers dissociation of species present) to the blast wave
22 equations.

23 An important consideration for a transition from a spark plasma of hot gas to a
24 self-sustained flame is the chemistry of ignition reactions. The chemical reactions
25 during this process can be divided in three steps [52]: chain initiating step, chain
26 propagating step, and chain terminating step. This transition begins with the
27 initiation of the chemical reactions and usually occurs in a few hundred
28 microseconds after the breakdown. During the ignition process, the second step
29 must be reached in such a manner as to obtain a self-sustaining flame. According to
30 the hot-gas ignition process, the hot-gas kernel must be sustained longer than the
31 initiation of the chain propagating step. This time corresponds to the ignition delay
32 of the flame, and is very sensitive to the mixture equivalence ratio, mixture type,

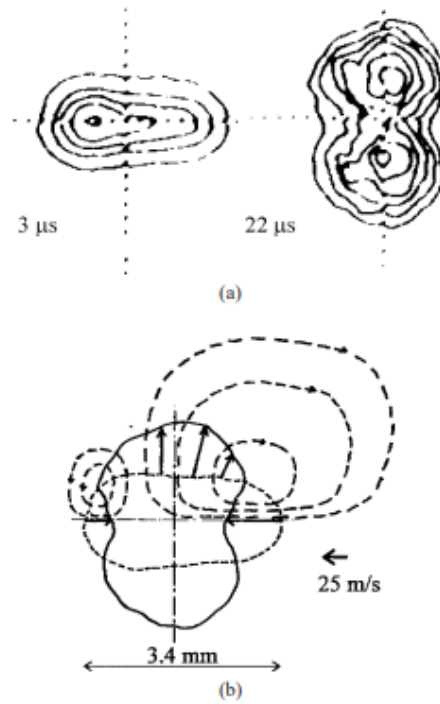
1 temperature, and pressure [83],[137]. The shape of the flame front during this
2 transition phase is called the flame kernel and is the focus of the present study.

3 **2.2.3.3 Dynamic features of the Laser-ignited Flame Kernel**

4 Based on photographic images of the early flame kernel acquired by different
5 research groups, the shape of the flame front at different time intervals reveals two
6 distinguishable propagation features. The first distinct feature is a toroidal flame
7 front propagating radially upward and downward in a direction perpendicular to
8 the laser beam axis. The toroidal shaped kernel is usually observed at the early
9 stages (i.e., $t < 100 \mu\text{s}$) and it is present in both laser and electric spark ignition.
10 Computer simulations by [138] shows that the toroidal shape of the kernel is due
11 to an induced inward flow which results from an over-expansion of the shock
12 wave, emanating outward from the region of the discharge at a very high velocity.
13 Figure 2.8a is a pair of LIF images captured at 3 and 22 μs showing transformation
14 from ellipsoidal spark kernel to a toroidal flame kernel [92]. When the two images
15 are superposed, a set of constant velocity streamlines can be drawn (Figure 2.8b)
16 which shows toroidal rings generated by the shock waves expansion. These are
17 indicated by the lengths of the arrowed vectors, the scale for which is indicated by
18 the arrowed line for 25 m/s.

19 The second distinct feature is a propagation of the flame front back towards the
20 ignition laser. The resulting shape is called the front lobe. The formation of the
21 front lobe follows after the toroidal flame kernel front is formed (i.e., $t > 100 \mu\text{s}$)
22 and it is peculiar only to laser ignition. There are different viewpoints on how the
23 front lobe is formed. One suggestion by [92] is that it may be due to the initial flow
24 field created by the propagation of a radiation transport wave up the laser beam,
25 arising from the high rate of energy transfer at the leading edge of the plasma.
26 However, it is argued that this propagation would cease shortly after the laser
27 pulse is ended ($\sim 15 \text{ ns}$ duration). Another explanation by [92] attributes the third
28 lobe to asymmetry to the spark with a corresponding asymmetry in the gas flow
29 that follows propagation of the shock wave and ensuing overexpansion. The
30 asymmetry of the spark is evidenced by the formation of an intense spark centre
31 located away from the midpoint of the spark volume, biased towards the spark

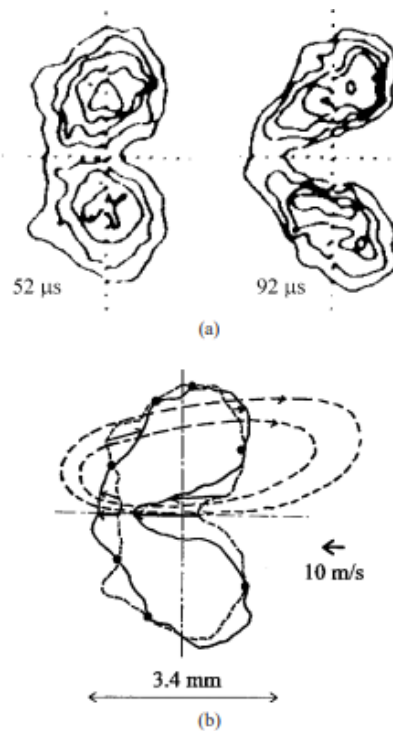
1 laser. The resulting shock wave will then propagate outward as discussed above
2 but with an axial component away from the intense spark centre and therefore
3 toward from the direction of the spark laser. Gases rushing to fill the hot, over-
4 expanded region then quickly ignite to form the kernels observed. A third
5 explanation by [20] is that the front lobe formation is due to the asymmetry in the
6 leading edge and trailing edge toroid that generates a centreline velocity towards
7 the laser source. The toroid generated by the rarefaction wave, as proposed in the
8 previous section, would appear capable of generating the lobe, particularly as the
9 toroidal ring at the leading edge would decay more rapidly. Figure 2.9a is a pair of
10 LIF images captured at 52 and 92 μs showing the early transformation leading to
11 appearance of front lobe [92]. By again superposing images, but now after a longer
12 elapsed time, it is possible to infer the direction and approximate magnitude of the
13 induced gas motion. After the weak toroid at the left has dissipated, the gas
14 velocity to the left, up the laser beam, can be high. This results in a flow of hot gas
15 close to the centreline to the left and its displacement by cold gas from the right.
16 The elongated hot gas kernel comprises the third lobe. It is always directed
17 towards the laser and is present in both non-reacting and reacting mixtures. Figure
18 2.10 represents the sequence of the flame kernel development in three-dimensions
19 which highlight key geometric features of the flame kernel.



1

2 **Figure 2.8: Formation of toroidal flame front: (a)LIF images showing transition**
3 **from spark to toroidal kernel (b) Streamlines showing toroidal rings in**
4 **superposed images [20].**

5



1

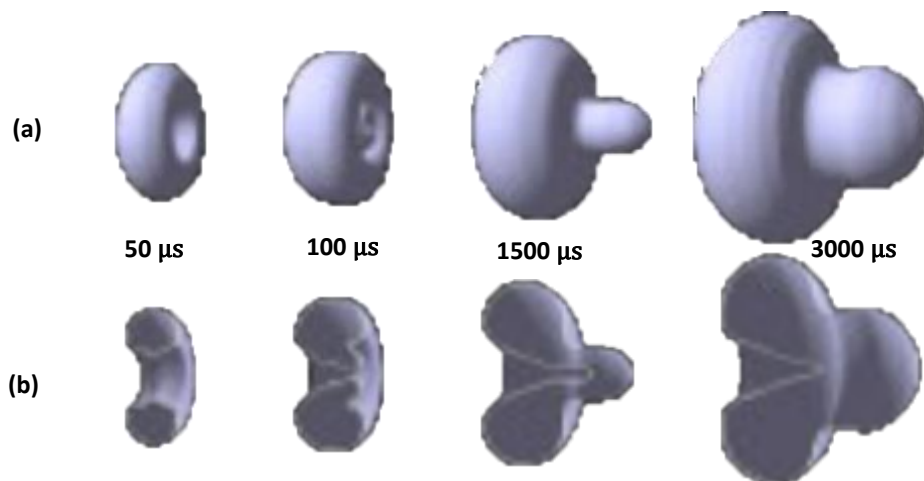
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Figure 2.9: Formation of the front lobe: (a) LIF images showing transition from spark to toroidal kernel (b) Streamlines showing toroidal rings in superposed images [20]

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Figure 2.10: Development of the Laser flame kernel: (a) Three-dimensional view, (b) Half-section view. [27]

8

9

10 The above features may not provide enough evidence for a successful ignition
 11 since the early kernel growth is mainly influenced by the gas dynamics of the shock
 12 wave and the high energy plasma. A necessary feature of an igniting kernel is the

1 production of sufficient quantity of radicals at the moment of chemical reaction
2 initiation. In [96], measurement of the temporal evolution of the OH* emission
3 intensity for two ignition trials (successful and misfiring cases), performed at the
4 same deposited energy, displayed an identical evolution during the first 100 μ s. In
5 this measurement, a peak in intensity corresponded to the generation and the
6 cooling of the spark. In the misfiring case, the intensity rapidly decreased to zero
7 after the peak, whereas in the firing case, this peak is followed by a second peak
8 between 400 and 1000 μ s, which corresponds to significant production of radicals
9 and thus to the initiation of the chemical reactions.

10 **2.3 Summary**

11 This chapter has provided insight into the theoretical basis for understanding
12 ignition initiation using a Laser beam and the propagation of the resulting flame
13 kernel in gaseous flow environment. It started with a discussion on the
14 fundamental properties of the flow as a foundation for understanding the
15 preparation of premixed flow environment for ignition to occur. Also discussed
16 are the fundamental theories of laser spark generation and the evolution to a
17 propagating flame which would form the basis to develop an understanding of the
18 flame kernel. Therefore, based on the gaps earlier identified from literature survey,
19 this report will further investigate the development characteristics of a LISI flame
20 kernel under premixed flow condition. The investigation will focus on the features
21 related to both the fluid dynamics and reactivity. In the next section, follows with a
22 description of the experimental methods employed.

Chapter 3 Experimental Setup and Measurement Techniques

This chapter gives an overview of the experimental facility and measurement techniques employed in the present study. The components of the experimental facility include the fuel and air supply system, mixture preparation and delivery system, atmospheric burners, laser ignition and various control/data gathering instrumentation. The main diagnostic techniques employed includes High Speed Laser tomography, High speed particle image velocimetry (HSPIV) and chemiluminescence imaging. Also, these experiments required detailed risk assessment and safe experimental procedures details of which can be found in appendices (see Appendix A: Experimental Procedures and Appendix B: Risk Assessment).

3.1 Fuels

Ignition and development of a flame requires the presence of fuel and oxidant mixture, either in premixed or non-premixed state. In all experiments, the fuel and oxidant were introduced as premixed charge before ignition. Three hydrocarbon fuel gases were investigated which includes: methane, propane, and ethylene. Table 3-1 is a summary of the of the three fuel gases investigated and their combustion properties.

Methane used for this study is technical grade CP methane, of 99.9% purity, supplied from a 200 bar BOC Gas Cylinder of 12 m³ capacity. It is a fuel gas with chemical formula, CH₄ consisting of a single carbon atom linked to four hydrogen atoms by single covalent bonds[139]. Due to its tetrahedral molecular structure, methane exhibits unique combustion characteristics which makes it an ideal fuel for experimental studies. For example, due to its large C-H bond energies, methane has a high ignition temperature and low laminar flame speed. In addition, it has unity Lewis number (*i.e.*, $Le = 1$) which means its data can be easily applied to

modelling validation by neglecting *Lewis number* effects. Above all, methane was chosen as the primary (i.e., base) fuel because it is the most widely researched, most understood and most widely implemented of all fuel gases[139]. The **Propane** used is commercial grade propane supplied from a 6 bar BOC Gas Cylinder of 11 kg weight. It is one of the liquefied petroleum gases (LPG) with chemical formula C_3H_8 . Propane is produced during natural gas processing and/or petroleum refining. It is economical to store and transport in liquefied form. Due its availability and adaptability as engine fuel, the use of propane as an alternative transport fuel is rising with about 17 million vehicles worldwide. The **Ethylene** used is a research grade ethylene supplied from an 80 bar BOC Gas Cylinder of 15 kg weight. This fuel gas has the chemical formula C_2H_4 consisting of two double-bonded carbon atoms each linked to two hydrogen atoms by single bond. It is a by-product of steam cracking of longer chain hydrocarbons (an energy intensive process that involved temperature of between 750-950°C). It is commonly used in agriculture to enhance the ripening of fruits and to stimulate rubber tree to yield more latex. Also used in petrochemical industry for polythene production by polymerization. Its thermos-physical properties compared to gasoline are strong indications that the gas can be easily combusted. For example, both the enthalpy of combustion (~ 49.46 MJ/kg) and autoignition temperature ($\sim 490^\circ\text{C}$) indicates better combustibility of the gaseous fuel in conventional gasoline in SI engines[8].

The oxidant used for all the fuels studied is **atmospheric air** supplied from the central laboratory compressed air system. It was chosen due to its established usage in combustion systems. The actual composition of dry air is 20.9% oxygen, 78.1% nitrogen, 0.9% argon, with trace amounts of carbon dioxide, helium, neon and hydrogen. However, these can be approximated to just 21% oxygen and 79% nitrogen, hence dry air is usually represented by the formula, $O_2 + 3.76N_2$ in chemical equations.

Table 3.1: Summary of fuel molecular and ignition properties

FUEL	Mol. wt. (kg/mol)	Density @ 20°C (kg/m ³)	Dyn. Visc @ 20°C (Pa. s)	Min Ign. Energy (mJ) [140]	LFL - UFL (vol%) [113]	max u_L @ s.t.p. (m/s) [112]	max T_A @ s.t.p (K) [110]
Methane	0.016	0.656	1.03E-05	3.00E-01	4.9 - 15.8	4.30E-01	2.23E+03
Propane	0.044	1.88	7.40E-06	2.60E-01	2.03 -10.0	4.40E-01	2.27E+03
Ethylene	0.028	1.26	9.60E-06	7.00E-02	2.74 - 31.5	6.60E-01	2.37E+03

3.2 Atmospheric Burners

For the experiments presented in this report, two different configurations of co-flow atmospheric burners were employed. The Co-flow burner is a Bunsen-type burner in which the flow mixture exits the system as *co-axial* gas jets. Similar to free gas jets, two distinct regions of flow can be spotted: a potential core region (where the jet velocity of the inner flow is maintained) and a mixing region (where the jet expands and interacts with the co-flow gas or surrounding air). The co-flow burner was chosen instead of a constant volume vessel because it provides complete optical access, allowing the use of any chosen laser diagnostic technique.

The first burner consists of two co-axial straight cylindrical tubes made from a 1.5 mm thick, Stainless Steel. This is the same burner used in [100] with an inner tube diameter of 10 mm and an outer (i.e. co-flow) tube diameter of 25 mm. In the initial study involving characterization of both the flow and flame regions using HSPIV, the reactants (i.e., methane and air) were externally premixed and admitted into the burner through a single inlet port. In the experiment, co-flow was not particularly required due to lower risk of flashback. A schematic of the setup used for HSPIV measurements is shown in Figure 3.1.

The second burner used for the chemiluminescence measurements is also an atmospheric co-flow burner but differs in geometry from the first burner. Unlike the first burner, it has a smoothly contoured nozzle with high contraction ratio. The inner and outer nozzles have exit diameters of 22 mm and 26 mm, respectively. Full details of the burner design can be found in [141]. In the

chemiluminescence study of the flame kernel development, the reactants were externally premixed and admitted into the burner via two opposite inlet ports. During the experiment, co-flow was required especially for the laminar flame cases due to high risk of flashback. A schematic of the setup used for the chemiluminescence measurements is shown in Figure 3.2.

3.3 Gas mixture preparation and delivery

Figure 3.1 is a schematic of the burner and mixture delivery setup used for the initial characterization by HSPIV technique. In this study, methane is the only fuel used. As shown in the figure, methane is supplied from a HP Gas Cylinder (200 bar BOC) fitted with a two-stage regulator. The air was taken from a central laboratory compressed air system, fitted with a pressure regulator (range: 0 - 6 bar). Gas delivery from both sources to the burner was via flexible piping and ball valves. The supplied methane was first metered by the digital mass flow meter (MV-304) and then passed through an electrically operated solenoid valve used to shut OFF and ON the fuel line from the control room. Also, the air was metered by a digital flow meter (MV-306) and then piped into a jet atomiser which contains olive oil to generate aerosol-air mixture for laser sheet flow visualization. Both flow meters have an accuracy of 1-2 % full scale. Finally, the metered methane and seeded air were premixed at a T-junction and then sent to the burner through a single inlet port.

Figure 3.2 is a schematic of the burner and mixture delivery setup used for the chemiluminescence measurements of flame kernel. In this study, all three fuel gases (methane, propane and ethylene) were used. All the reactants were prepared in similar fashion as above except that the metered air did not require seeding. After the reactants were premixed at a T-junction, the mixture was passed through a flashback arrestor to prevent back-flow and then sent to the burner through two opposite inlet ports.

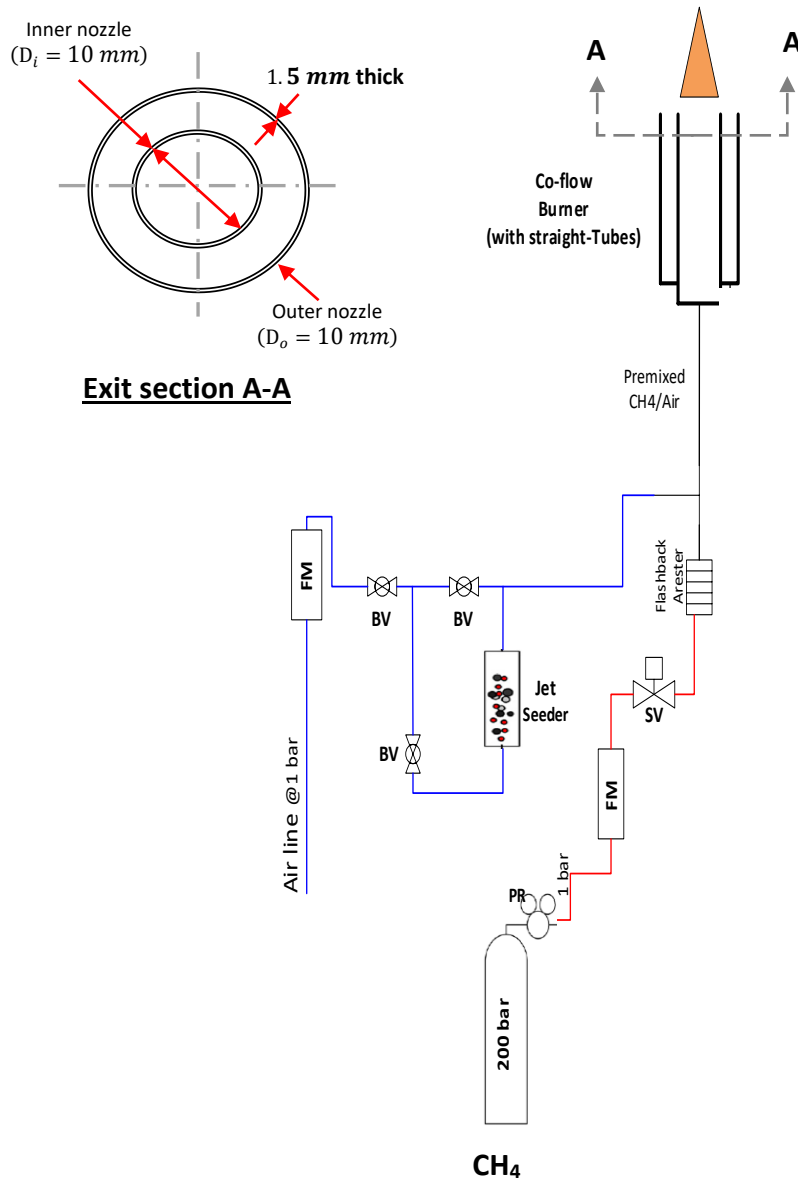


Figure 3.1: Schematic drawing of Burner, Flow Setup used for HSPIV.

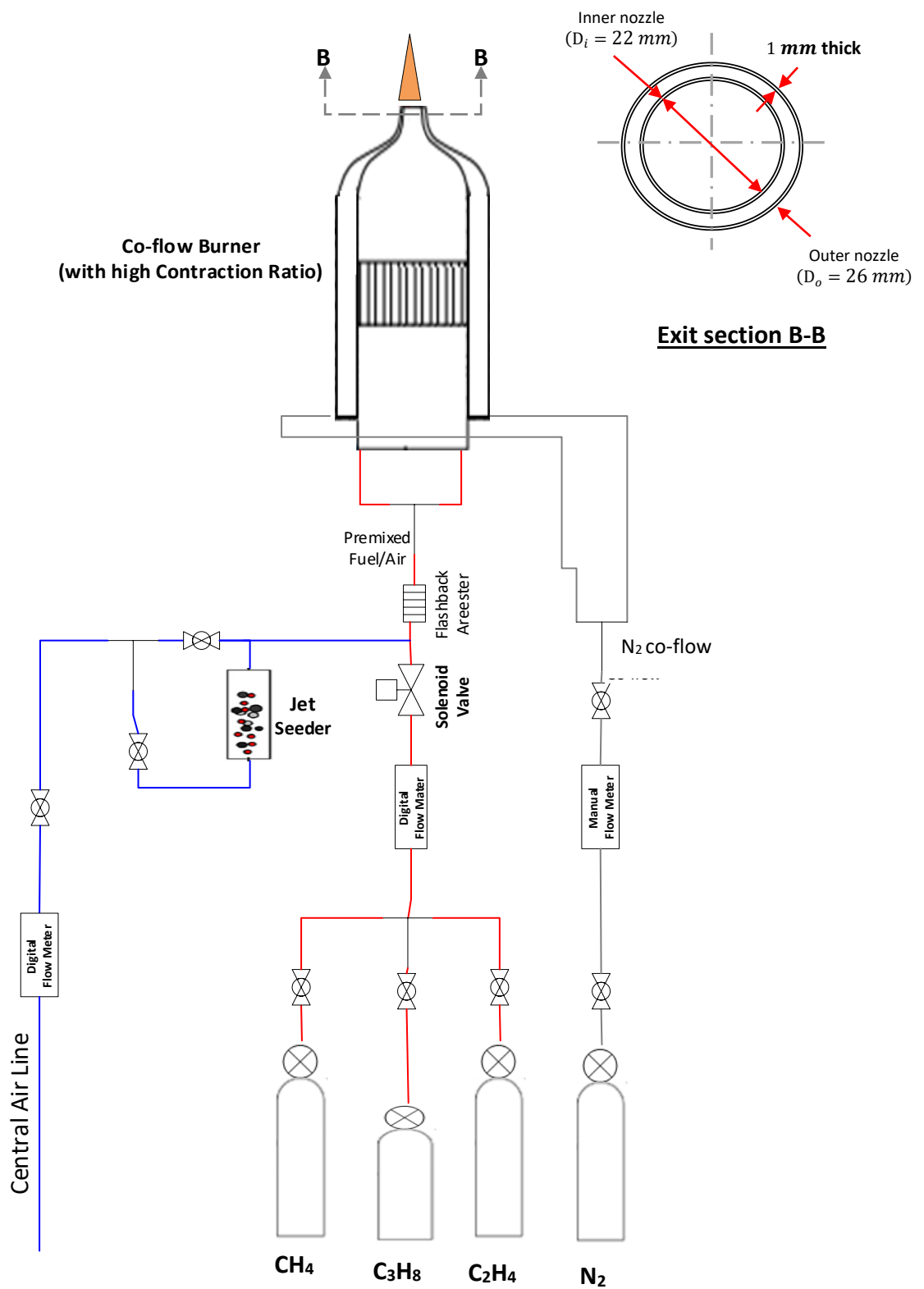


Figure 3.2: Schematic Drawing of Burner, Flow Setup used for Chemiluminescence.

3.4 Instrumentation and Measurement Techniques

3.4.1 Laser ignition, pulse energy and spark spot size

Laser ignition: For all tests, the ignition of premixed fuel gas-air charge was achieved using the spark generated from a Q-switched Nd:YAG laser beam (*Continuum, Surelite III*) which is capable of delivering 500 mJ maximum pulse energy and 7 ns pulse duration (at Full Width Half Maximum) at its fundamental wavelength of 1064 nm. The output laser beam at fundamental wavelength was steered through 90° by a right-angle triangular prism and then focussed at 20 mm height above the Burner centre using a Plano-Convex lens of 75 mm focal length and 25 mm diameter. This height was chosen to ensure the spark occurred within the potential core region of the gas jet issuing from the burner, as revealed by the velocity profiles obtained from the PIV. Also, it allows sufficient space for development of the flame-kernel. To ensure that each ignition event is time-independent, the laser Q-switch was operated at a pulse rate of 1 or 2 Hz while the flash lamp was operated at its design frequency of 10 Hz.

Pulse energy: Before each ignition, the laser output power per pulse was measured using a pyroelectric laser energy sensor and display meter (*Gentec, UNO*). The energy output is controlled by changing the delay in timing of the laser Q-switch from the flash lamp output. Figure 3.3 is a typical energy profile of the laser at different Q-switch delays. For most of the conditions investigated, a constant energy of 32 mJ was obtained using Q-switch delay 530 μs. This energy is higher than the required minimum ignition energy (MIE) at the selected equivalence ratios of the different fuel-air mixtures.

Beam profile and spark spot size: Using a burn paper, the spot marks produced by the laser beam shows an approximately Gaussian beam profile while the focused beam has a size of about 1 mm diameter. The beam profile gives information about the beam quality. Both parameters are important because they affect the minimum energy required for plasma formation and ignition. For a Gaussian beam profile, the beam quality (defined by the M^2 factor) is related to the spot size at its focal point according to the equation:

$$\text{spark spot size } (2r) = \left(\frac{4f\lambda}{\pi D} M^2 \right) \quad 3.2$$

where:

- D input beam diameter at the lens (at the 1/e² point),
- M^2 Beam mode parameter ($M^2 > 1$ except for a perfectly Gaussian beam)
- λ Laser wavelength
- f Focal length

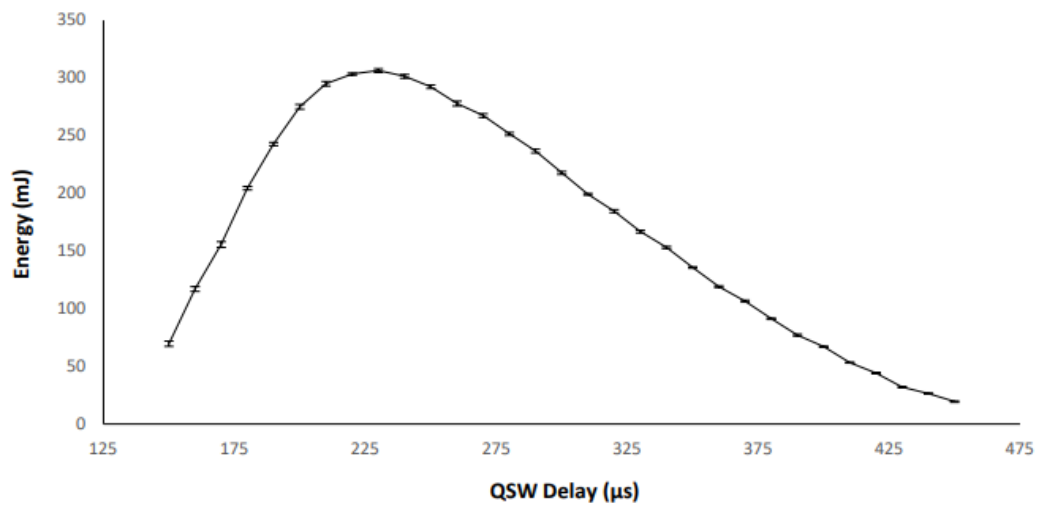


Figure 3.3: Typical Energy profile of the Ignition Laser

3.4.2 Laser sheet flow visualization and velocity field measurement

Characterization of the ignited flame kernel and its flow field was achieved using a combination of high-speed laser sheet tomography and particle image velocimetry (PIV) techniques. Laser sheet tomography enabled visualisation of the and further extraction of the flame kernel characteristics while PIV processing was employed to show the flow field structures such as the velocity vectors and turbulence intensities. The system arrangement used to deploy both techniques is the same.

3.4.2.1 Particle image velocimetry

Particle image velocimetry is a non-intrusive laser diagnostic technique, which provides instantaneous velocity vectors of the flow field. The principle of PIV is based on recording of two separate images with time delay between them as

shown in Figure 3.4. Usually seed particles (or traces) are used to seed airflow, which reflect light when illuminated by a laser pulse. The flow seeded with particles is then illuminated twice and two images are recorded with small time separation between them. The velocity vectors are derived through measurement of the particle movement as shown by the acquired images in the two interrogation windows.

The interrogation windows from each image frame are cross correlated with each other. A typically interrogation window has a dimension of 16×16 or 32×32 pixels[142]. The cross-correlation procedure is repeated for each interrogation window over the two images captured by the camera producing a signal peak in each interrogation window. A sub-pixel interpolation is used to measure the velocity with high accuracy. The velocity computation is directly dependent on a number of particles in the flow. It is advised to have at least 10 to 20 particles per interrogation window[143].

The accuracy of the measurements is dependent on the ability of the particles to follow the flow and adjust their velocity to the flow fluctuations. If the flow is subjected to extreme acceleration, for example just behind the flame front, the inertia of the particles can affect the velocity measurements. This effect can be crucial for application of PIV to combustion [144].

3.4.2.2 Laser tomography

Laser tomography is similar to PIV in the sense that it relies on the illumination of tracer particles seeded in the flow. However, the objective of this technique is not to obtain velocity vectors but to obtain a plane cross section of the flame and to visualize the shape of the flame front [145]. Usually, an oil aerosol is injected into the unburned mixture, and a laser beam formed into a thin sheet illuminates the combustion zone. The oil particles scatter the laser light in the unburned region and thus the beam can be visualised, but in the area where the flame had passed the oil would have evaporated and burned, so do not scatter the light.

A limitation of this technique is that it only allows visualisation of the two-dimensional section of the flow field. Therefore, visualisation of three-dimensional structures is unavailable. The primary use of this technique is for visualisation

purposes, however secondary quantitative data can be gathered from each frame of the recordings. Using High Speed Laser tomography for flame propagation studies, secondary data on the flame arrival time, flame position and therefore flame speed can be obtained.

The errors involved in this data can result from the collection of high-speed images. Magnification effects of the lens used on the camera can lead to errors in the exact definition of the region of interest. Errors in flame arrival time can also result from delays in the triggering of the recording system.

3.4.2.3 Optical Setup

Figure 3.5 is the optical arrangement used to simultaneously visualize the flow and the flame kernel in a two-dimensional plane. It includes seeded flow, illumination source, beam delivery optics, signal synchronization box and a high-speed camera.

Flow seeding

For both PIV and tomography measurements, air was flowed through a jet atomiser containing olive oil to generate the seeded flow. An important consideration in the selection of seeding material is that the particles must trace the flow with sufficient fidelity to accurately follow the important turbulent structures. For accurate measurement, typical particle diameter required for olive oil seeding is between 0.98 to 3.09 μm [146].

Flow Illumination

An important requirement for the seeding is that individual particles of the material must scatter enough light to allow definition and recording of the flow movement. Based on the assumption of circular symmetry of the particle, the theory of light scattering states that the intensity of scattered light depends both on two main characteristics: the size of the particle, and wavelength of the incident light. Based on these characteristics, measurement of light scattering intensity has been classified into three distinct regimes which include: Rayleigh regime, Geometric optics regime and Mie scattering regime. In Rayleigh scattering [147], the radius of the particle is much smaller than the wavelength of the incident beam. This results in a very small light scattering intensity and therefore is not suitable for imaging low

velocity flows [148]. In Geometric scattering, the particle radius is much greater than beam wavelength. For such particles, the solutions present the use of a large number of terms, which in order to calculate requires the use of a computer. For the study present in this thesis, Mie scattering [149] is the chosen technique. The technique involves the imaging of light scattered by droplet particles whose radius is comparable to the wavelength of the incident beam [149]. In the present setup, illumination of the oil-seeded flow is achieved by a dual pulsed Nd:YLF laser (Litron, LDY304 model), each with wavelength 527 nm, maximum power 30.5 W @ 1 kHz. The PIV laser was mounted on the flat surface of a movable table which was rigidly clamped to the floor to prevent any motion or vibration. For all measurements, the laser pulse setting was 1.3 kHz frequency and 55% energy (corresponding to 15.8 W for laser 1 and 16.1 W for Laser 2).

Sheet formation

The beam from PIV laser was directed to a sheet forming optics device (TSI, 610026) using a flexible light arm (TSI, 1098915 model), with knuckles capable of rotating 360°. Light sheets of 527 nm wavelength exiting the optics were focused on the region of interest in the flow, illuminating the olive oil seeded airflow. The sheet optics device was mounted on an inclined optical bench to prevent any likely damage from the ignition laser beam.

Imaging system

Mie-scattered images of the seeded flow were recorded using a High speed TSI Camera (630106 model, 36 gb memory). The Camera has a resolution of 1 Megapixel (1280 x 800), a pixel size of 20 um x 20 um and a frame rate of 7400 at full pixel resolution. The camera was positioned so that the image plane was perpendicular to the plane of the illuminated flow sheet. A Micro NIKKOR lens ($d = 55$ mm, $f = 200$ mm) was fitted to the camera to provide imaging and was operated with an aperture setting of $f/32$. To filter out chemiluminescence from both plasma and flame, a 527 nm laser-line filter (50mm Diameter, 20 nm Bandwidth) was placed in front of the lens.

Trigger timing and synchronization

Both the laser source and the camera were synchronized using the Synchronizer (TSI, 610036 model) whose function is to control the firing of the laser source and camera frame capturing. A typical timing sequence of the PIV setup used to characterize the flow is shown in Figure 3.6. For the flame kernel development study, both the ignition and the PIV system were initially triggered using the same TTL signal from a *Stanford*, DG535 Function Generator. The PIV software was designed to allow a camera trigger delay time to be set with respect to the external input signal. However, this feature malfunctioned during trial tests, so a TG5011 Delay Generator was included to delay the PIV firing/capture from the ignition.

Software implementation and data acquisition

The Insight software allowed on-screen control of the camera and laser for accurate timing and data acquisition. The software interface is shown in Figure 3.7. The orange triangle with dashed line represents the energy profile of the laser beam which was obtained using varying Q-switch delays. The on-screen window allowed setting of laser pulse separation, Δt and the required camera trigger delay, allowing adjustments to the amount of light scattered by the seeding particles for a given camera aperture. For visualization of the flow and flame kernel, the laser pulse repetition rate was set to 6 kHz equivalent to 166 μs total PIV exposure time (or 83 μs per frame for a double-frame CCD camera). The delay from the initial camera synchroniser TTL trigger to the firing of the first laser was set to 75 μs , while a laser pulse separation, 50 μs was applied to allow the capture of the velocity field range in two separate frames. The captured image from the ignition test has an area of 384 by 800 pixel (i.e., 19.2 mm by 40 mm).

The images obtained were stored as TIFF files, which were later processed to obtain flow velocity and flame tomographic information.

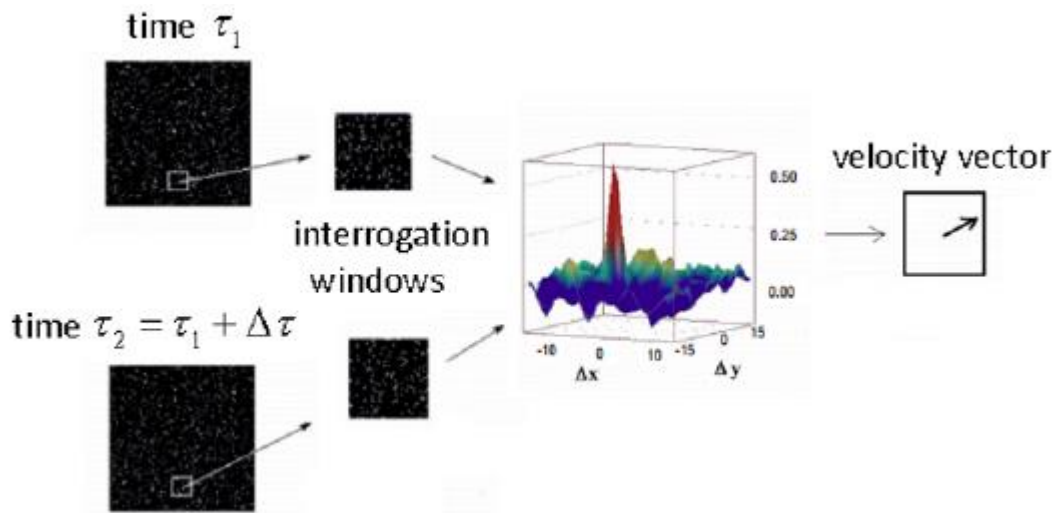


Figure 3.4: PIV cross-correlation technique[150]

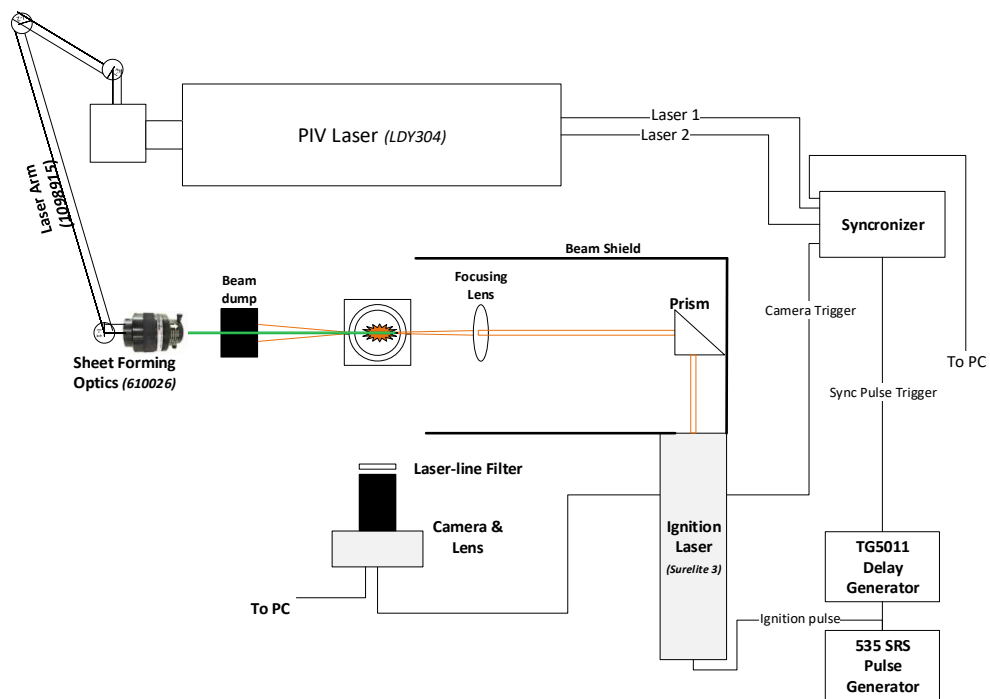


Figure 3.5: Optical Layout for PIV and Tomography Measurements

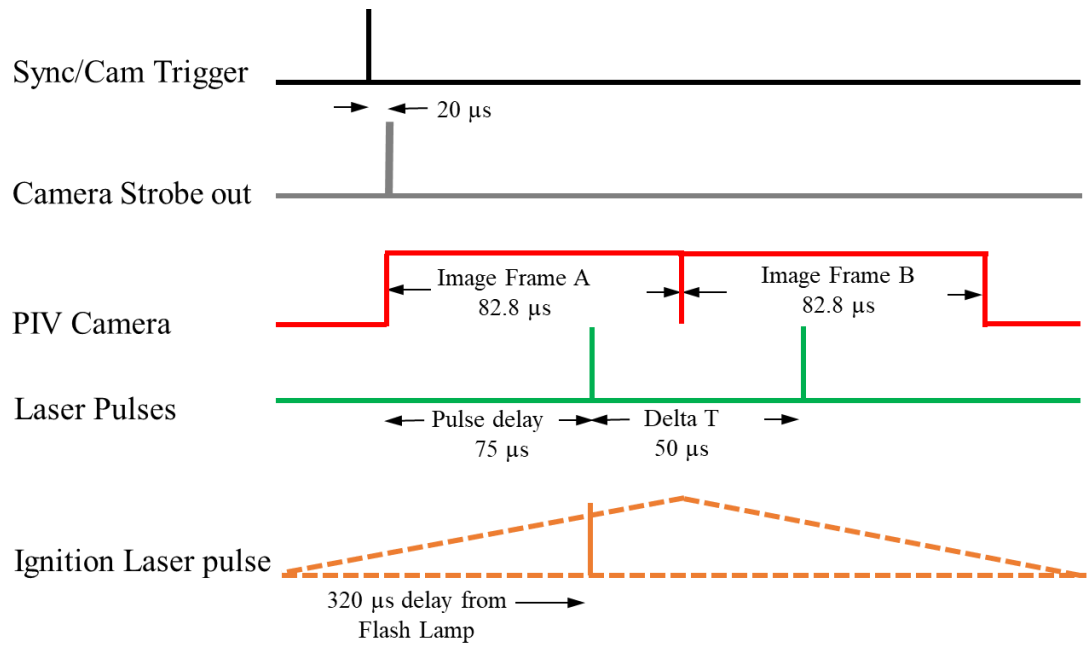


Figure 3.6: Timing Sequence for PIV and Tomography Measurements

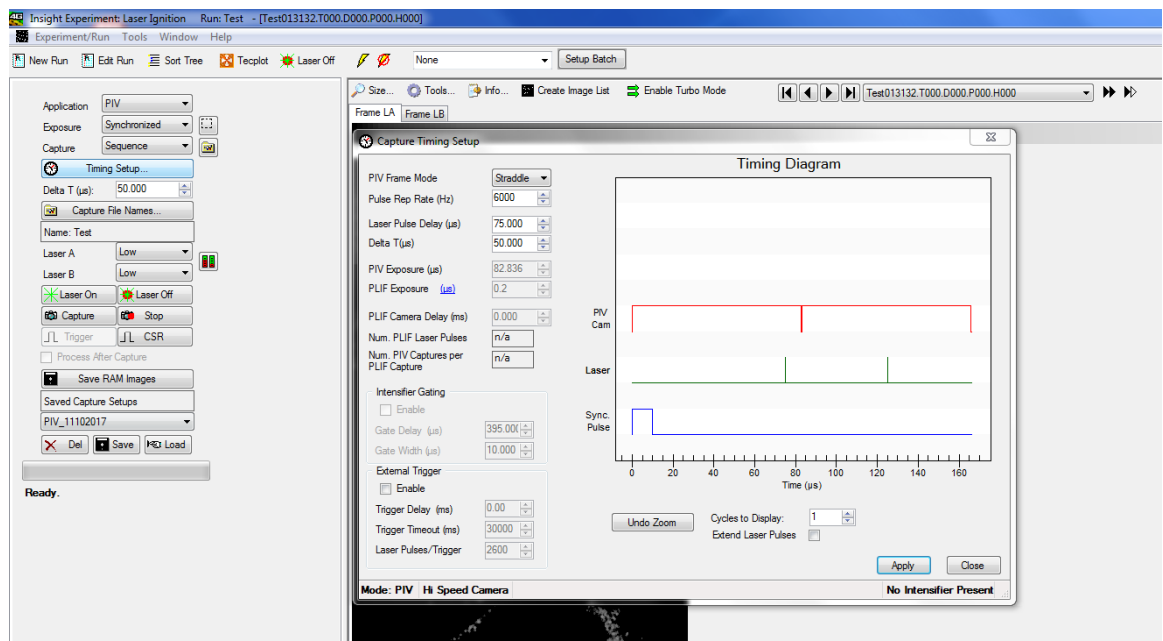


Figure 3.7: PIV Software (Insight 4G) Interface

3.4.3 OH* Chemiluminescence Imaging

Chemiluminescence imaging was performed mainly to capture the characteristics of the flame kernel and its dynamics. Chemiluminescence involves capturing the radiations emitted by electronically excited molecules in the flame, while returning to their lower energy states. From emission spectroscopy, it is long known that the radiation wavelength is a characteristic of a particular molecule and the transition the molecule undergoes. As shown in Figure 3.8, the hydroxyl radical (OH*) is one of the most used markers of flame presence in combustion studies because its emission spectrum exhibits major narrowband wave peak around 308.9 nm wavelength. The OH* is an intermediate species which is formed by fast two-body reactions, such as the attack of CH radical on O₂ molecules. Unlike diffusion flames where the OH* tends to occur in thin, tendril-like structures (indicative of reactions occurring at stoichiometric fuel/air interface), OH* appearance in premixed flames can exist over a large spatial region (commensurate with the extent of the reaction zone). Hence, it is a good flame marker for the present study.

Figure 3.9 is a schematic of the experimental setup used for the chemiluminescence studies reported chapters 5-7. OH* chemiluminescence signals from the developing flame kernels were captured using a 16-bit intensified CCD camera (*Princeton Instruments*, P-MAX II) fitted with a UV Camera Lens (78 mm diameter, f/3.8). The camera uses a CCD array size of 512 x 512 pixel, and its spectral range is 180-900 nm, hence it is sensitive in both ultraviolet and visible regions. In order to capture the OH* chemiluminescence, a combination of UG 11 and WG 305 Schott glass filters were placed in front of the UV lens to filter unwanted radiations.

The camera was externally triggered using the laser output signal, **fixed sync out**, which is the same as the Q-Switch signal used for laser ignition. The timing sequence is shown in Figure 3.10. During this test, the laser Q-Switch which was internally triggered and operated at 2 Hz frequency. All camera settings used for data acquisition were remotely controlled via the commercial PC software (*Princeton*, Winspec32) provided by the camera manufacturer. The software platform enabled camera gate width and delay to be set. In all the experiment

conducted, a constant gate width of 20 μs was used while ignition delay was varied between 50 μs and 10000 μs . The software also allowed captured images to be saved either as individual TIFF files or in matrix SPE file format. For each delay, a total of 200 images were captured and each capture image has an area of 512 by 512 pixel (i.e., 94.72 mm by 94.72 mm). These images were later processed with codes written in MATLAB to obtain both geometric and radical intensity information of the flame kernel.

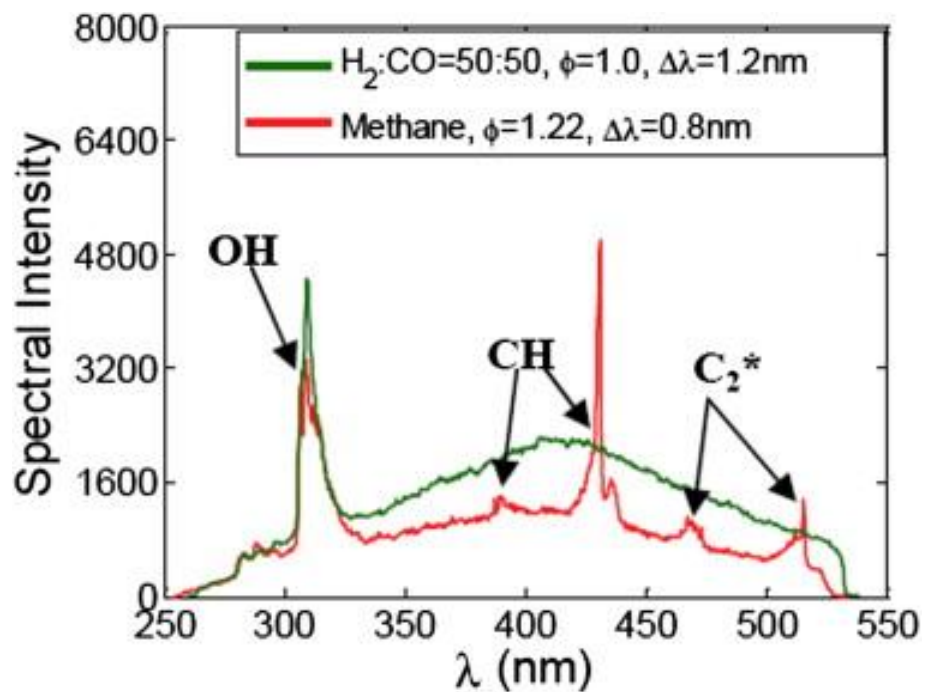


Figure 3.8: Typical flame spectra in syngas and methane flames [66].

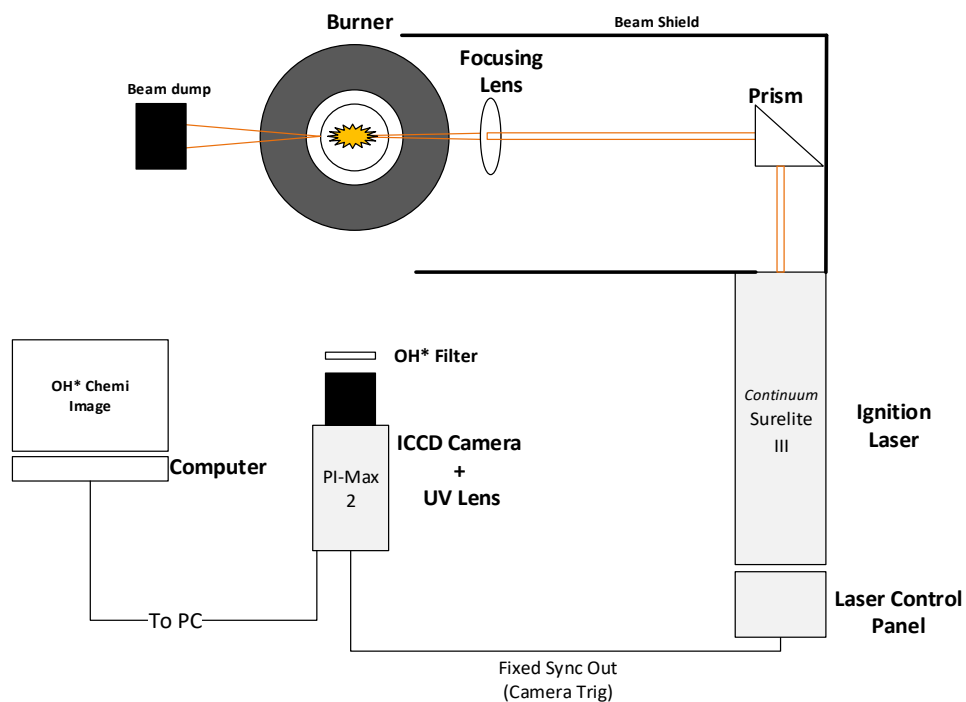


Figure 3.9: Optical Layout for Chemiluminescence Imaging

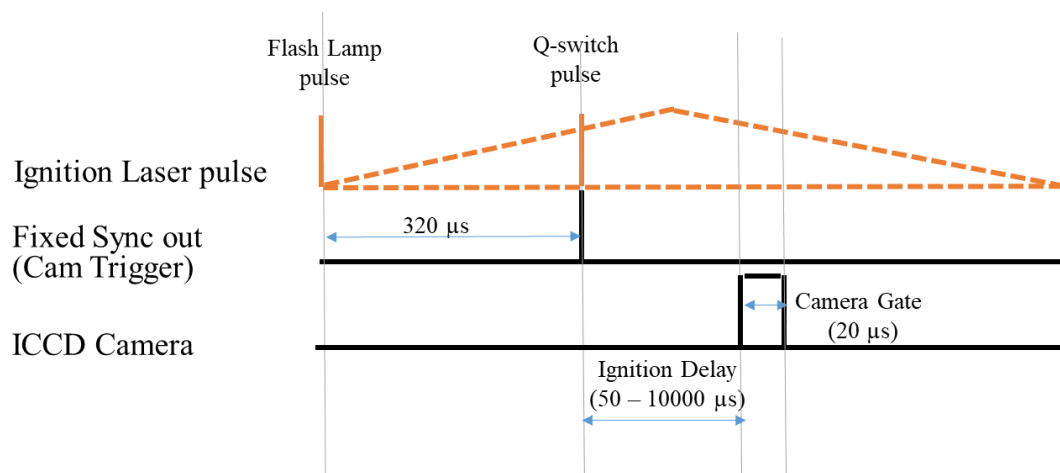


Figure 3.10: Timing sequence used for Chemiluminescence Imaging.

3.5 Data Processing

3.5.1 Processing of Velocity field data

Prior to the measurements, a target image of known dimensions was obtained. Using the calibration function present in the software, the x and y scale was input to allow conversion from pixel to mm. The calibration values obtained were 52 μm per pixel (x scale) by 52 μm per pixel (y scale).

Once the images were captured, the raw image were **pre-processed** with the Insight 4G software. The first pre-processing step involves subtraction of a minimum background image (generated by the Insight 4G software) from the raw images. Due to non-uniformity of the two image frames, background subtraction was done using two pre-processors with the first pre-processor generating the background image and the second subtracting the background image from each image. This was useful for enhanced image quality (by reducing signal-to-noise ratio) and for obtaining a uniform image for the PIV. The second pre-processing step involved masking of the background subtracted image. This was done to separate the flame boundary from the flow. To enable movement of the boundary, a dynamic mask was created by setting the intensity threshold value as one. Also, the images were conditioned to restore intensity in flow areas with low intensities.

After the initial image pre-processing, velocity vectors of the flow were obtained using in-built functions provided in the software. Insight 4G performs velocity field calculation using a two-frame cross-correlation routine, which incorporated an FFT correlation engine and a Gaussian peak search algorithm. This correlation method differs from autocorrelation and single-frame cross-correlation, in that it achieves image particle separation by recording the first laser pulse images on frame 1 and the second laser pulse images on frame 2, thus allowing the velocity to be measured without directional ambiguity. By repeated application of the cross-correlation routine to the interrogation window pairs (Figure 3.4), an intensity spectrum is obtained from which particle displacement and velocity were computed.

For the current analysis, a final grid size of 32 by 32 pixels (i.e., interrogation region) with a maximum displacement limit of 4 pixels. This grid size proved to give good results with reasonable resolution compared to grid sizes of 64 by 64 pixel. No masking was used in processing the results. This is due to the high computational time required when masking is used for processing. The default FFT correlation engine was used due to its speed. The Gaussian peak correlation is the default setting with FFT, and thus it was selected. Insight 4G allows the user to specify the minimum number of particles and the signal to noise ratio of the interrogation spot required for a velocity vector to pass. The defaults of 10 particles per interrogation spot, and a signal to noise ratio of 1.5 were chosen. Table 3.2 lists the settings used for PIV images processing using Insight.

The cross-correlation method of processing PIV images can generally produce over 95% correct velocity vectors [151], however, spurious vectors can also be produced. For the results obtained during this study, good agreement was achieved with the calculated bulk flow velocity, however, as with all experimental investigations using PIV, a small number of false vectors were apparent. Insight 4G provides a number of inbuilt macros, ranging from a standard deviation filter to a smooth filter, which allowed the validation of the computed results. Results presented in this thesis were subjected to a median filter. The local median filter applied, compared each velocity vector in the field with a median value calculated from the surrounding neighbourhood vectors. Full details of the validation scheme can be found in [152].

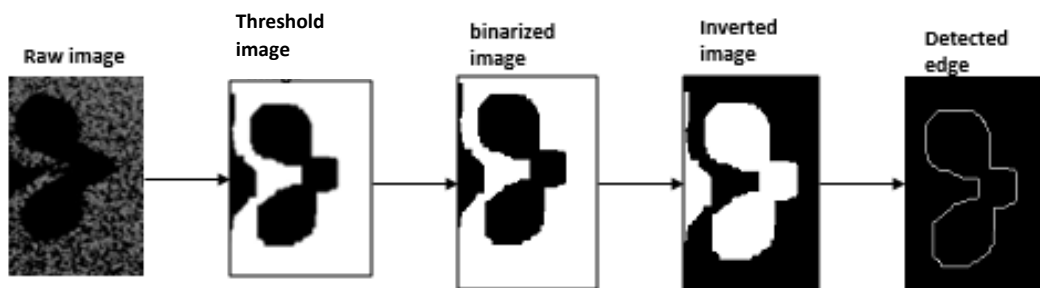
Finally, the results of the processed data were exported in excel format. A code written in MATLAB was further used in visualising the flow velocity vectors as well as the turbulent intensities. Detail of the code are contained in section C3 of Appendix C

Table 3.2: PIV Processing Settings

Grid Engine	Nyquist	Grid size	32 x 32
Mask Engine	No mask	Signal-to-Noise Ratio	1.5
Correlation Engine	FFT	Max. Displ (pixel)	4
Peak Engine	Gaussian	Min. # of particles	10

3.5.2 Processing of flame kernel data from the Raw Tomographic Image

Following the initial pre-processing of captured imaged using Insight 4G software, further processing was done to extract the flame boundary using MATLAB. Figure 3.11 shows the processing steps from the raw to the boundary detected edge of the flame kernel. After pre-processing, the first processing step in MATLAB was binarization and image inversion. To be able to separate the flame region from the flow, it was necessary to create a binary (i.e., black and white) image, where the white part is the flame, and the black part is the flow. This was achieved using two MATLAB functions (a binarization function followed by an inversion function). Finally, the flame edges were detected using another MATLAB function (known as canny edge detection). From the detected edges, the geometric properties of the flame kernel were obtained. The codes written for these processing is contained in section C1-C2 of Appendix C.

**Figure 3.11: Image processing steps**

3.5.3 Processing of Flame kernel data from the OH* chemiluminescence Images.

Following acquisition of chemiluminescence images, computer codes written in MATLAB were used to process the images (see Appendix D: MATLAB Code Used to Analyse Chemiluminescence Data). Figure 3.12 shows the main processing steps. The first processing step involved averaging of the 200 images acquired for each ignition delay. This step was necessary to reduce shot-to-shot variation in the analysed data and it formed the basis for further analysis.

To obtain the flame geometric characteristics, image thresholding and binarization which was performed on the mean projection images to distinguish the flame area from the fresh gases. This was achieved by first applying a multilevel thresholding algorithm based on Otsu's thresholding technique [153]. Next, a binarization function was applied. To smoothen the flame edges, a median filter was applied. Finally, the boundary of flame edges was extracted based on an edge detection algorithm.

From the mean projection image, both qualitative and quantitative data were extracted. The visualisation quality of the images was improved by introducing colour scales as shown by the colour-mapped image. Due to the wide range of data, all the images could not be plotted on the same scale, however comparisons could be made based on the spatial structure and intensity scale of individual images.

Also, quantitative values of the spatially integrated intensity and peak intensity were directly extracted. To investigate the uncertainties in the selected sample size both the integrated intensities and peak intensities were compared for different sample sizes at different time delays. Figure 3.13 shows the normalised distribution of integrated OH* intensities with respect to sample size at various time delays which shows a variation range between 0-1% of the maximum value. Likewise, the normalised distribution of peak OH* intensities with sample sizes shows a variation between 0-5% of the maximum value as shown by Figure 3.14.

From the extracted edges, key geometric information about the flame front locations and propagation rate were obtained.

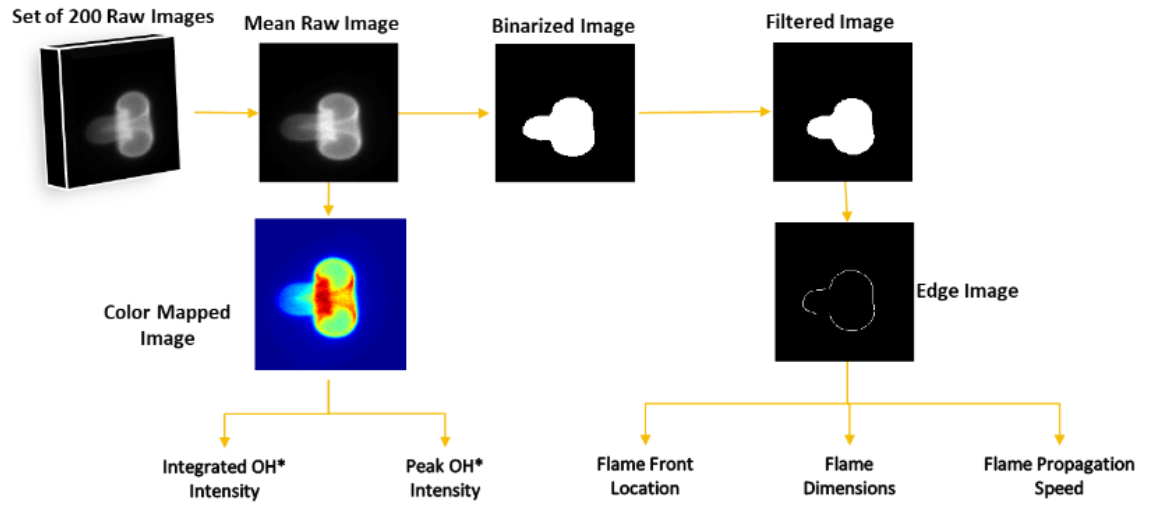


Figure 3.12: Chemiluminescence Image processing steps

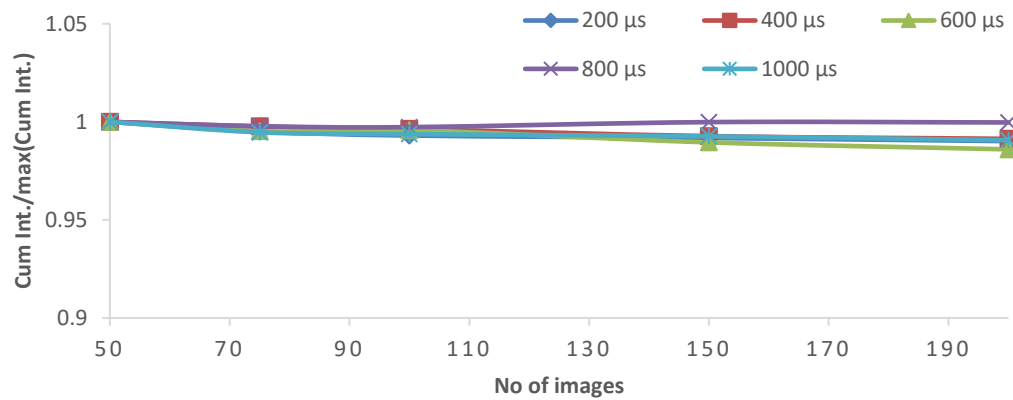


Figure 3.13: Normalised distribution of Integrated OH* intensity w.r.t sample size.

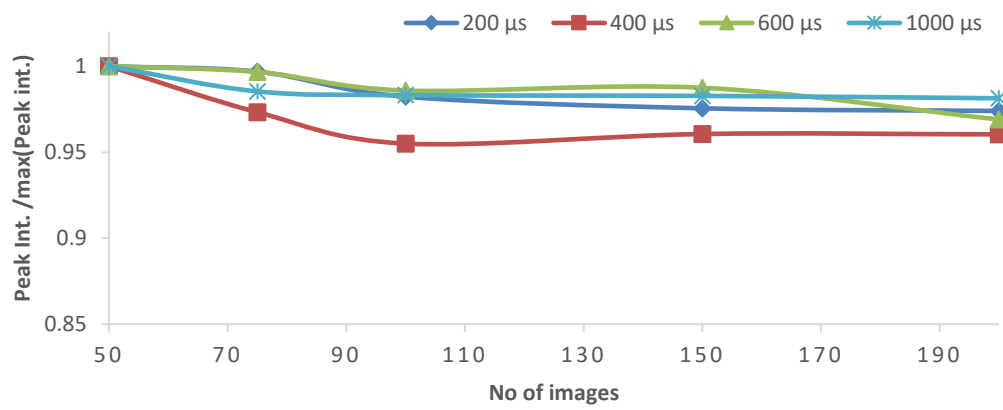


Figure 3.14: Normalised distribution of Peak OH* intensity w.r.t sample size.

1 3.6 Summary

2 This chapter presented the experimental setup and measurement techniques used
3 in this work. The measurements were conducted based on two experimental
4 setups and three optical techniques developed during this PhD. The first setup
5 consists of a co-axial straight tube burner in which air-seeded methane was flowed
6 and illuminated by using a double-pulsed PIV Laser. Following ignition of the
7 mixture, high speed tomographic imaging of the flow field was achieved by
8 filtering out the flame chemiluminescence using a laser-line filter. Analysis of the
9 tomographic images would allow simultaneous investigation of the flame kernel
10 features and its flow field. The second setup consists of co-axial contracting nozzles
11 burner with in which different fuel/air mixtures were flowed. Repeated laser
12 ignition and subsequent capture of the natural OH* chemiluminescence of the
13 flame kernel enable analysis of both the fluid dynamics and reactivity of the flame.

1 Chapter 4 Flame Kernel and Flow-field Characterisation of 2 Laser-Ignited Methane-Air Mixture

3 4.1 Introduction

4 The flame kernel represents the transition phase between the initiation of
5 chemical reactions in a flammable mixture and a fully developed or self-sustained
6 flame. In the early stage of laser ignition, measurements of the flame kernel
7 characteristics are particularly important because the expansion speed of the
8 flame kernel is low compared with that of a fully-developed flame [41],[98]. For
9 example, the flame kernel shapes and sizes are useful for predicting the pressure
10 rise with time inside a combustor which maybe too small to be detected by existing
11 pressure probes. These properties are also useful for estimating the burning
12 velocity which is a fundamental parameter required for the development and
13 validation of kinetic mechanisms for fuels. One of the advantages offered by laser
14 ignition is flexible choice of ignition location. To optimize combustion based on
15 ignition location, there is a further need to characterise the properties of local flow
16 field prior to ignition and during the ignition event. There have been numerous
17 studies relating to the fluid dynamics of the laser-ignited flame kernel
18 [92],[134],[20],[154], however there is a lack of data on the effect of the kernel
19 expansion on surrounding flow field structure. In addition to optimizing
20 combustion based on ignition location, such data is important for understanding
21 the interaction between flame generated turbulence and the fluid shear generated
22 turbulence.

23 In this chapter, high-speed images obtained from the 2D-flow sheet of laser-ignited
24 stoichiometric Methane/Air mixture was used to characterise the flame kernel and
25 its flow field. The study is divided into two main sections. In the first section, the
26 characteristics of the laser flame kernel was qualitatively described based on
27 tomographic images obtained using Laser sheet flow visualization. Further

1 extraction of the flame kernel edges enabled quantitative description of the kernel
2 characteristics such as the flame front location, kernel size and growth rate. The
3 second section begins with characterisation of the burner under cold isothermal
4 conditions. Finally, the flow field properties in the reacting state is described.
5 Visualisation of the flow in both isothermal and reacting state was based on PIV
6 technique from which the flow field quantities like velocity and turbulence
7 intensity were extracted.

8 **4.2 Flow conditions**

9 To characterise the development of the flame kernel and its flow field, two flow
10 conditions of stoichiometric methane-air mixture were selected. The flow
11 conditions were selected based on the Reynolds number of the flows (one laminar
12 flow case and one turbulent flow case). A summary of the test conditions for the
13 two test cases is shown in Table 4.1: Summary of flow conditions. **Methane (CH₄)**
14 was chosen because of its unique combustion characteristics. For example, its high
15 ignition temperature and low laminar flame speed makes it an ideal fuel for
16 experimental studies; It has unity Lewis number (i.e., $Le = 1$) which means its data
17 can be easily applied to modelling validation by neglecting Lewis number effects; It
18 is the most widely researched, most understood and most widely implemented of
19 all fuel gases. In addition, methane is the main component of Compressed Natural
20 Gas (CNG) which is regarded as one of the most promising alternative fuels for
21 major combustion systems. **Atmospheric air** was chosen as the oxidant due to its
22 established usage in combustion systems. Dry atmospheric air is composed of
23 20.9% oxygen, 78.1% nitrogen, 0.9% argon, with trace amounts of carbon dioxide,
24 helium, neon and hydrogen. However, these can be approximated to just 21%
25 oxygen and 79% nitrogen, hence dry air is usually represented by the formula, O_2
26 + 3.76 N_2 in chemical equations.

27

28

1 **Table 4.1: Summary of flow conditions**

Test case #	1 (Laminar)	2 (Turbulent)
Equivalence Ratio, ϕ	1.0	1.0
Air Flow Rate (litres/min)	23.8	100.95
CH ₄ Flow Rate (litres/min)	2.5	10.6
Bulk flow velocity, U_0 (m/s)	1.15	4.89
Flow Reynolds Number, Re (Pa. s)	1694.35	7184.03
Adiabatic flame temperature, T_{ad} (K)	2226	2226
Laminar flame velocity, S_L (cm/s)	41	41

2

3 **4.3 Flame kernel Characteristics**4 **4.3.1 Visualisation of Flame Kernel Development**

5 This section discusses visual characteristics of the propagating flame kernel based
6 on high-speed images acquired using two-dimensional Laser tomography. Figure
7 4.1a is a sequence of images showing the time evolution of the instantaneous flame
8 kernel obtained for the laminar case while Figure 4.1b shows the evolution for the
9 turbulent case. As shown by the figures, the first frame of both test cases contains
10 an *elliptically shaped* high intensity plasma (usually observed within the first 1-3
11 μ s) which is usually accompanied with shock waves. As observed by many authors
12 [92],[20][100], in the early stage the plasma usually extends more towards the
13 direction of laser beam and the shockwave centroid is usually displaced from that
14 of the plasma. Its initial size can be inferred from the mechanism of electrical
15 breakdown and energy transfer from the laser beam.

16 The second common feature of both tests observed in frame 2 (about 50 μ s later)
17 is a *toroidal-shaped kernel* with two lobes propagating radially upwards and
18 downwards. As observed in the past [20], the toroidal shaped kernel is the results
19 of two contra-rotating vortices (A and B) of different strengths which are

1 generated through interaction of the shock waves and rarefaction waves from the
2 hot plasma. An illustration of the transformation from ellipsoidal spark kernel to a
3 toroidal flame kernel had been explained earlier using superposed images
4 captured between 3 μs and 22 μs (see Figure 2.8a & Figure 2.8b). As earlier
5 described, the inward flow resulting from these vortices leads to the formation of
6 the toroidal flame front. Computer modelling by [138] had shown the generation of
7 the toroidal flame front during simulation of the discharge from an electric ignition
8 source, thus this feature is not unique to laser ignition.

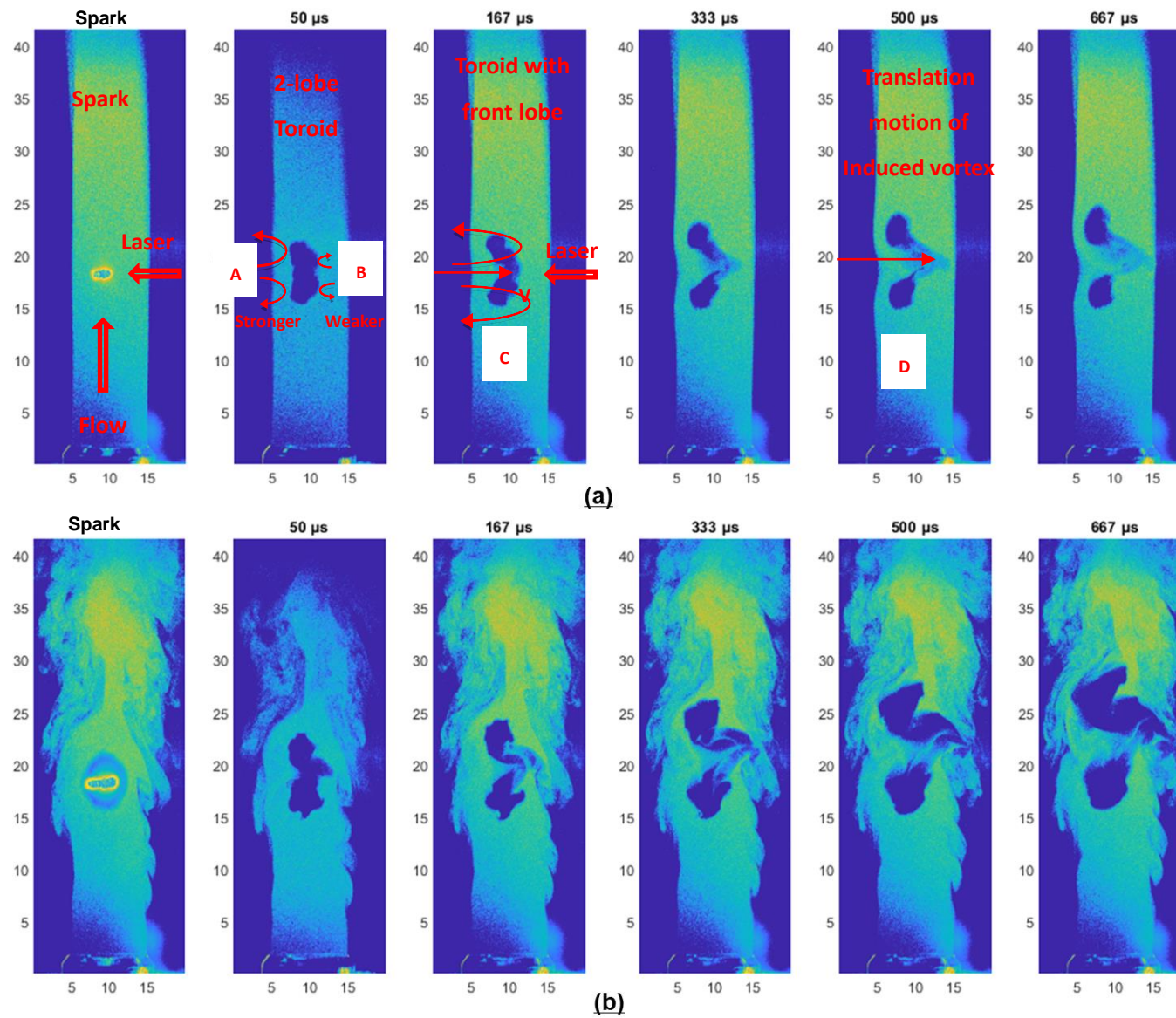
9 A third common feature observed at 167 μs , is the appearance of a third feature in
10 frames 3, known as *front lobe* which propagates in the direction of the laser as
11 shown in subsequent frames. The generation of the third lobe has been attributed
12 to many factors by past authors. For example, Spiglanin *et al.*, 1995 [92] suggested
13 that it might be due to the initial flow field created by the propagation of radiation
14 transport wave up the laser beam, due to the high rate of energy transfer at the
15 leading edge (i.e., upstream) of the plasma. While it is true that the plasma kernel
16 created by laser breakdown can result in an ionized front propagating toward the
17 laser [155],[20] this is not expected to continue long after the laser pulse had
18 ceased, hence, there is no evidence of this being the source of the front lobe
19 feature.

20 As earlier observe in the toroidal kernel, the two contra-rotating vortices (A and B)
21 are of different strength with A, growing stronger and B fast decaying. As
22 suggested in [20], this phenomenon is capable of producing a centreline velocity
23 directed towards the laser source, leading to generation of the front lobe (as
24 shown by C). Figure 4.3 clearly illustrates this transformation in the superposed
25 kernel edges formed between 51 and 168 μs . As shown in the figure, after the weak
26 leading-edge toroid has dissipated, the induced gas velocity moves to the right
27 towards the laser beam. This results in a flow of hot gas close to the centreline to
28 the left and its displacement by cold gas from the right. The elongated hot gas
29 kernel comprises the third lobe. Overall, the evolution of the kernel shape is
30 similar for both laminar and turbulent flow cases, however there is large
31 differences in the number and magnitude of local curvatures at each ignition
32 delays due to the extent of interaction with the flow field. The three-dimensional

1 geometric features of the flame kernel have earlier been illustrated (see Figure
2 2.10).

3 With respect to the kernel interaction with the surrounding flow field, it can be
4 seen that for the laminar case, the initial kernel shape is unaffected by the flow in
5 the first 168 μs . As previously stated, the appearance of the front lobe results from
6 the asymmetry in the upstream and downstream toroid rings which generates a
7 centreline velocity towards the laser source. This can be observed in the impinging
8 jet flow moving from left to right (as shown by D) towards the laser as the kernel
9 grows. By 668 μs , the jet catches up with the flame front referred to as the front
10 lobe enabling mixing of the upstream and downstream flows originally separated
11 by the flame kernel. This mixing could lead to many scenarios. The first is local
12 quenching which is observed around the front lobe where stretch effects are
13 usually high. The second is a splitting of the flame kernel into two combustion
14 zones as observed in subsequent images. In addition, the kernel becomes distorted
15 in shape due to interaction with the flow. A better picture of the flow-field effect on
16 the flame kernel is observed in the velocity vector plots.

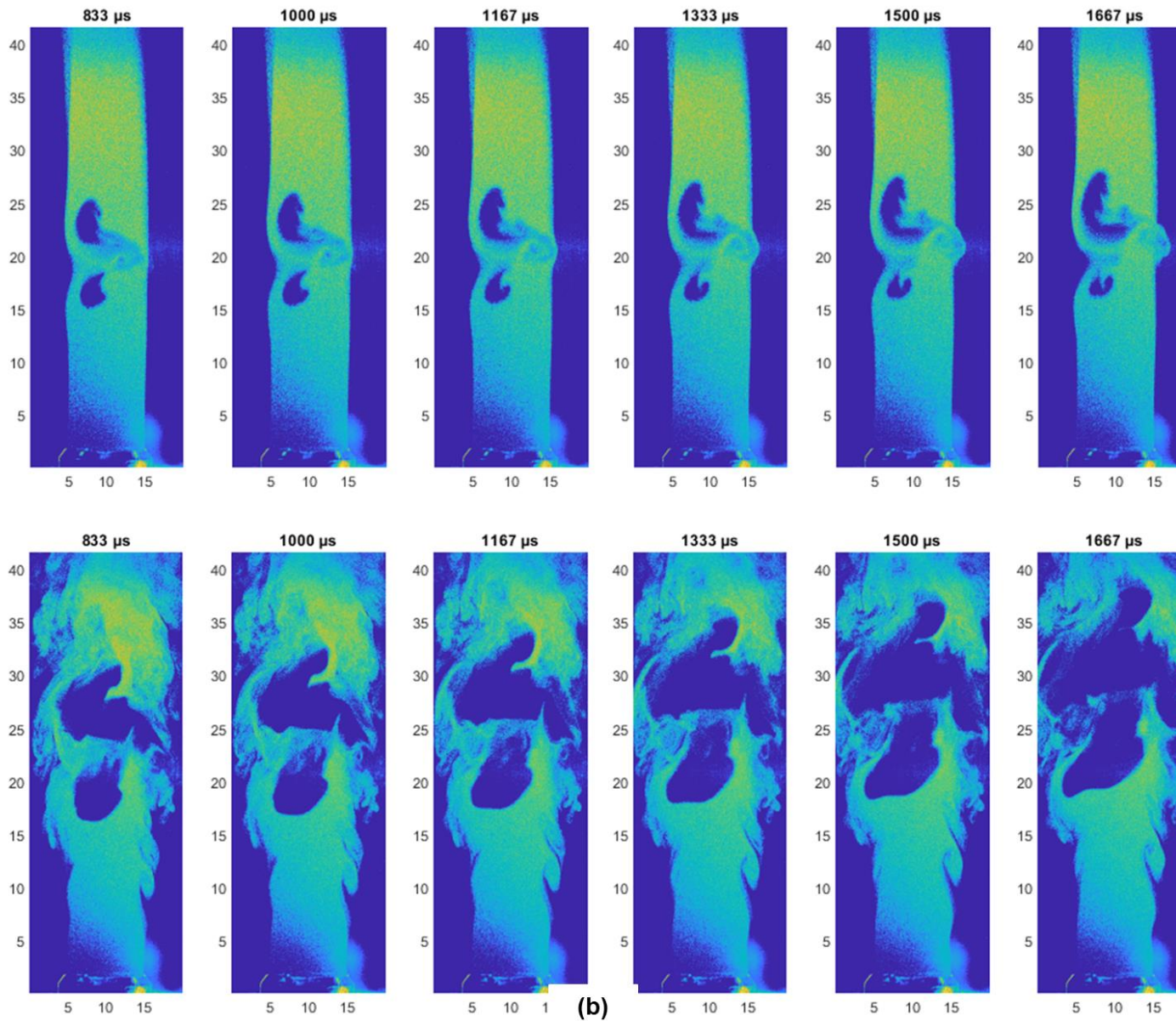
17



1

2

Figure 4.1: Flame Kernel Evolution in: (a) laminar CH_4/Air , and (b) turbulent CH_4/Air



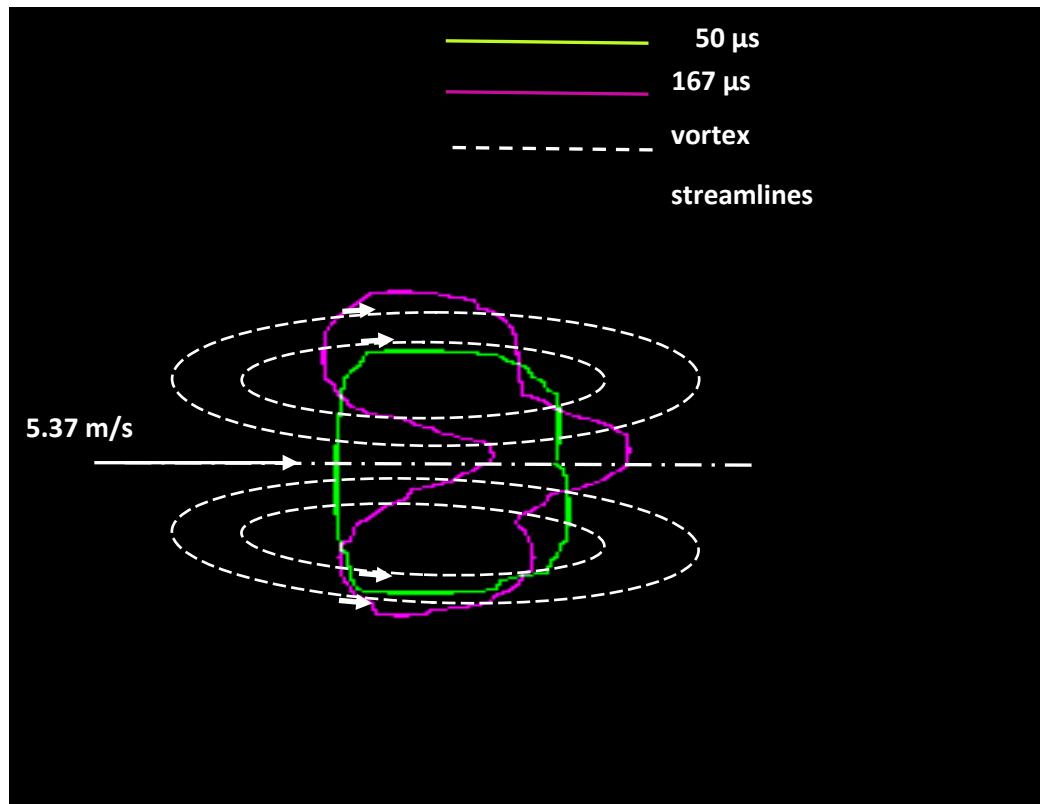
1

2

Figure 4.2: Flame Kernel Evolution in: (a) laminar CH_4/Air , and (b) turbulent CH_4/Air {continued}

1

2



3

4

Figure 4.3: Illustration of Front lobe formation

5

1 4.3.2 Characterisation of the Flame Kernel

2 Based on the high-speed images obtained using two-dimensional Laser
 3 tomography, geometric characteristics of individual flame kernels were extracted.
 4 The first characteristics investigated was the initial size of the plasma for both test
 5 cases. As observed in [90], there's nearly an instantaneous growth of the plasma to
 6 an initial finite size due to sudden energy deposition at breakdown which
 7 generates high pressure in the kernel leading to a rapidly expanding wave. From
 8 the equivalent circles of the extracted edges of the initial plasma kernels (i.e. 1 us
 9 delay) in Figure 4.4 and Figure 4.5, the initial plasma radii were found to be 1.57
 10 mm for the laminar case and 1.90 mm for the turbulent case. In Table 4.2,
 11 comparison is made between the measured values and those predicted using
 12 Taylor's Blast wave model (see equation 2.15) assuming 70% spark efficiency.
 13 Based on the comparison, the measured radius of the initial plasma for the laminar
 14 case is approximately 13.9% less than the predicted radius while that of the
 15 turbulent case is 4% more. The observed differences in the initial sizes of the
 16 kernel for both conditions despite having similar pulse energy highlights the fact
 17 that laser induced gas breakdown is a stochastic process, hence similar pulse
 18 energies can produce different spark energies, although the same spark efficiency
 19 of 70% has been assumed in the prediction.

20 **Table 4.2: Measured versus predicted initial plasma radius.**

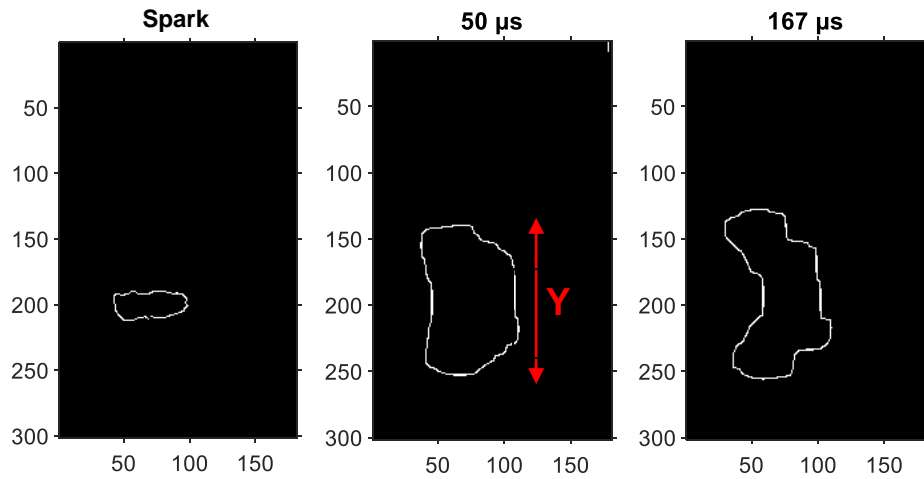
Test Case	Measured R (mm)	Predicted R (mm)
1. Laminar	1.57	1.82
2. Turbulent	1.90	1.82

21

22 Following determination of the initial sizes of the plasma, the evolution of the
 23 flame kernels was characterised based on the *kernel sizes* and *growth rate* at each
 24 delay. This time, the kernel size was quantified based on the maximum
 25 displacement along the vertical Y-direction (i.e., the flow axis) as shown by Figure
 26 4.4 and Figure 4.5. Figure 4.6 and Figure 4.7 shows the respective sizes and

1 locations (i.e., upstream, and downstream locations) of the flame front along the
2 axis of the flow for both laminar and turbulent flow conditions. As observed in
3 both plots, the initial development of the kernel is marked by rapid growth (i.e., in
4 the first 50 μs), however the growth rate declines before 200 μs . For example, in
5 the laminar flow case, the kernel size increased by 360% (i.e., 1.3 mm to 6 mm) in
6 the 50 μs and by further 8% in 167 μs , while that of the turbulent flow grows from
7 1.5 mm to 7.5 mm in 50 μs (i.e., 400% increase) and a further 23% increase in 167
8 μs . This initial rapid growth is mainly due to the gas dynamics of the decaying
9 plasma (i.e., independent of heat release due to chemical reaction) and gives little
10 or no indication of a successful ignition. The large decrease in growth rate by 200
11 μs shows the dwindling influence of the plasma gas which could mean the end of
12 kernel life for non-igniting mixtures. The second phase of kernel development
13 occurred between 200 μs and 400 μs due to the high stretching of the front lobe.
14 The third phase which starts around 400 μs indicates the end of chemical
15 induction process and the start of actual ignition. During this stage kernel growth
16 is influenced by a combination of gas dynamics and chemical reaction. Around 1.2
17 ms the influence of plasma gas dynamics is non-existent as kernel growth is mainly
18 as a result of heat release due to chemical reaction. A clearer picture of these
19 stages and the extent of influence of both processes (i.e., plasma gas dynamics and
20 chemical reaction) is shown by the growth rate plots in Figure 4.8 and Figure 4.10.
21 The general trend in growth rate involves reduction from an initial high value to an
22 asymptotic value close to S_L . From the normalized growth rate versus
23 displacement (Figure 4.9 and Figure 4.11), it can be seen that the growth rate
24 drops from an initial stretched value of $1.7S_L$ to about $0.2S_L$ having moved a
25 distance of 10 mm. A similar trend is observed in the turbulent case, however
26 unlike the laminar case there was more fluctuations in growth rate (especially
27 beyond 1 ms) due to turbulence influence.

28



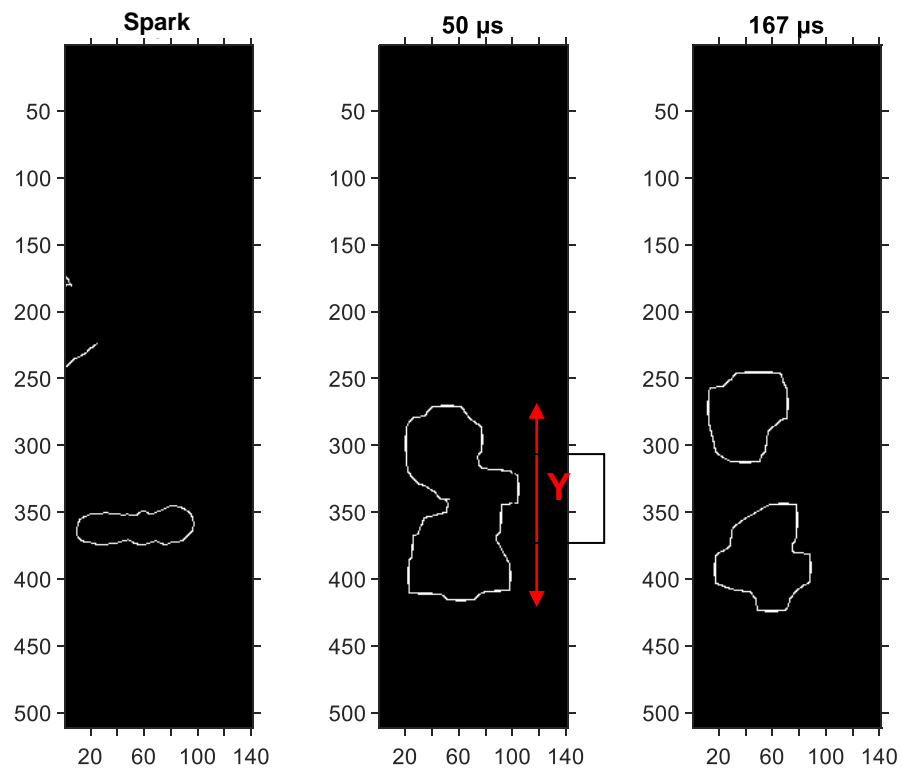
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Figure 4.4: Extracted Edges of Flame Kernel in Laminar Flow; Y = max. displacement along flow axis.

3

4

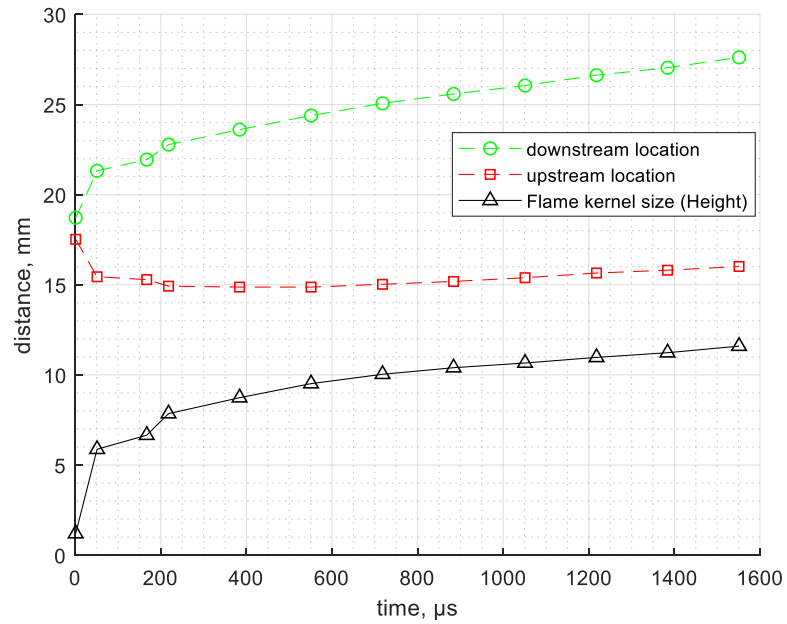


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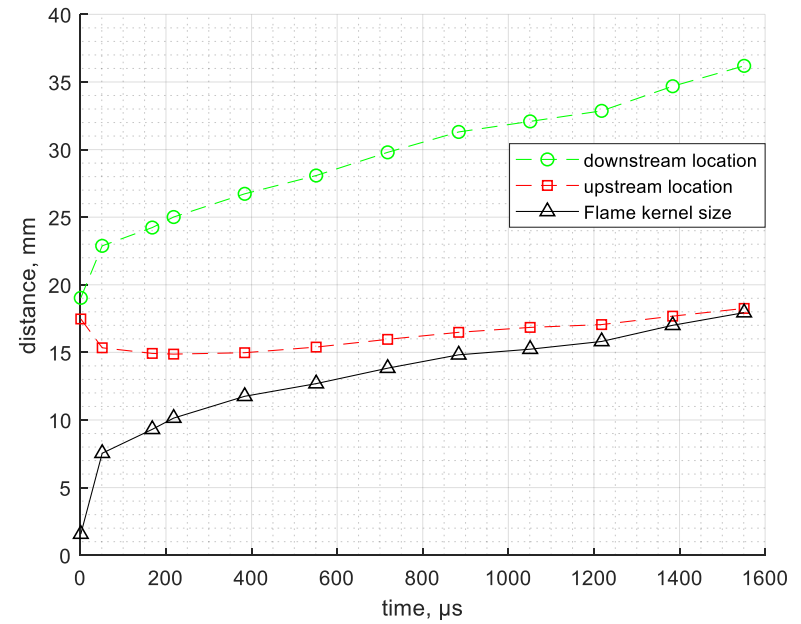
Figure 4.5: Extracted Edges of Flame Kernel in Turbulent Flow; Y = maximum displacement along the flow axis.

7



1

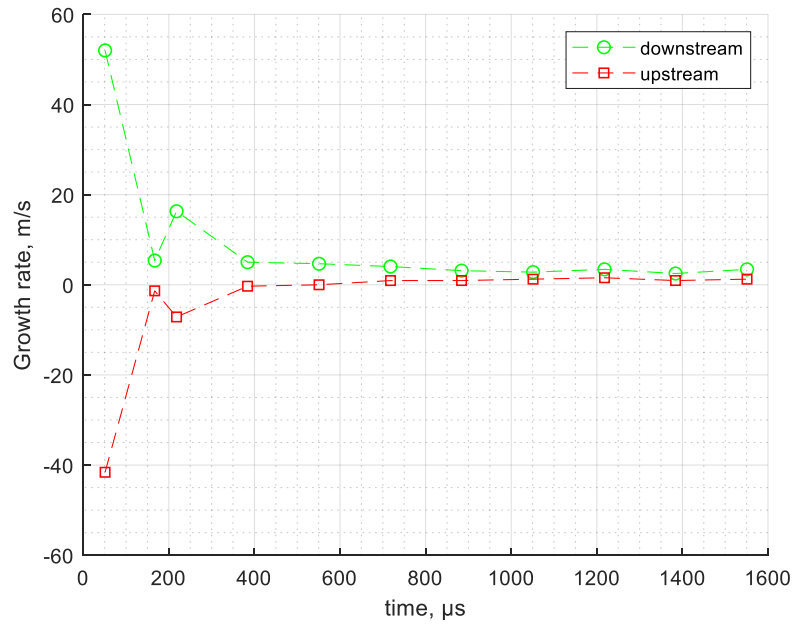
2 **Figure 4.6: Axial Flame Kernel Propagation (Laminar case)**



3

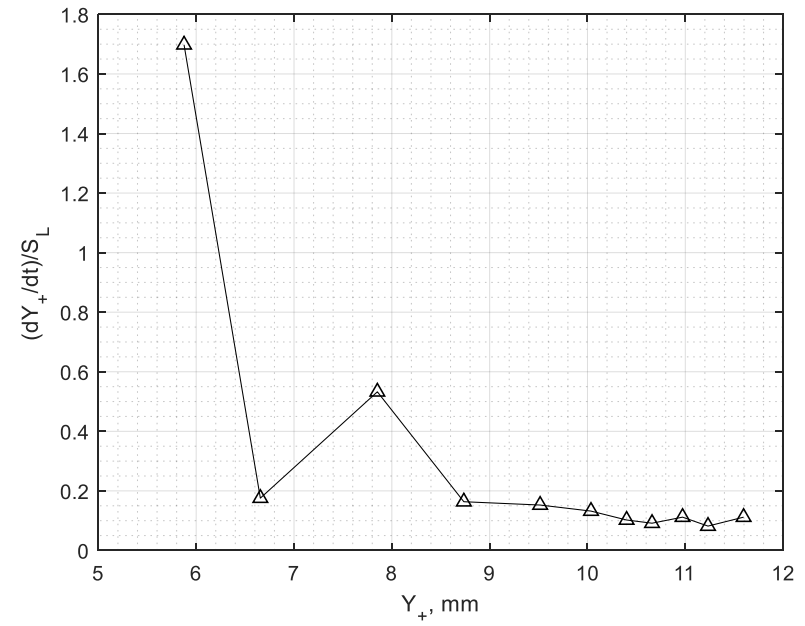
4 **Figure 4.7: Axial Flame Kernel Propagation (Turbulent case)**

5



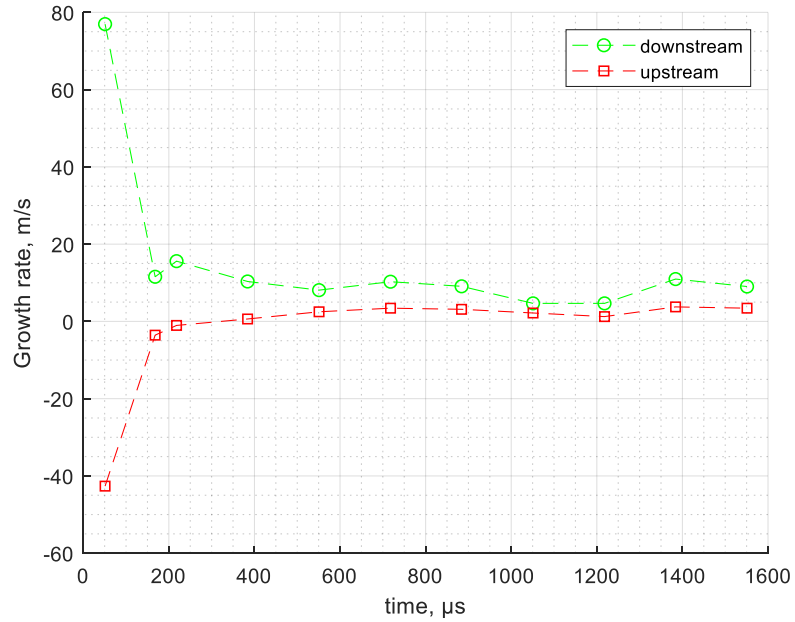
1

2 **Figure 4.8: Axial growth rate of Kernel (Laminar)**



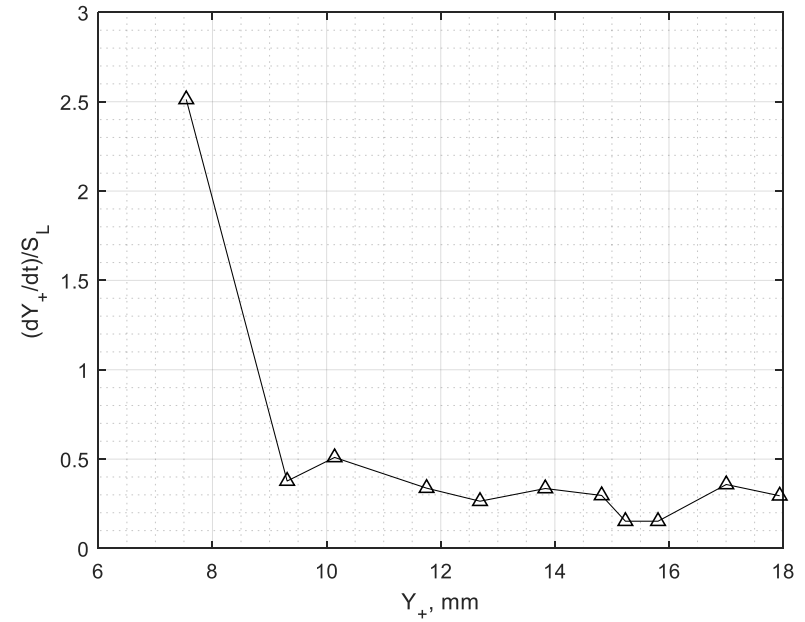
3

4 **Figure 4.9: Normalised Downstream Growth rate (Laminar)**



1

2 **Figure 4.10: Axial growth rate of Kernel (Turbulent)**



3

4 **Figure 4.11: Normalised Downstream Growth rate (Turbulent)**

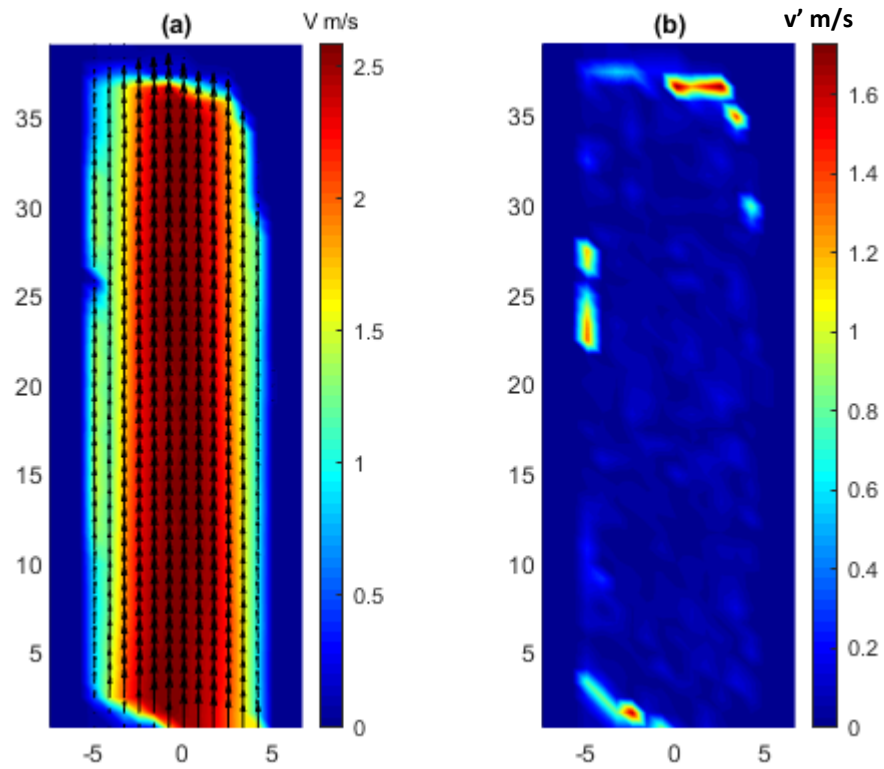
1 **4.4 Flow-field Characteristics**

2 **4.4.1 Characteristics of the Isothermal cold flow**

3 Figure 4.12a - Figure 4.13a shows the mean velocity vector plots for the laminar
4 isothermal and turbulent isothermal flows whose calculated bulk velocities were
5 1.15 and 4.89 m/s respectively. As expected, both exhibits a jet-like flow structure
6 with peak velocities in the potential core area which depreciates as you move
7 radially outwards. The jet profile for the laminar flow (Figure 4.14) reveal an
8 axisymmetric parabolic shaped flow structure the velocity along the centreline jet
9 remains nearly constant at all downstream location. In the turbulent flow situation
10 (Figure 4.15), the jet structure is similar, however, the centreline velocity
11 decreased with height. This is due to the jet expansion and interaction with the
12 surrounding air.

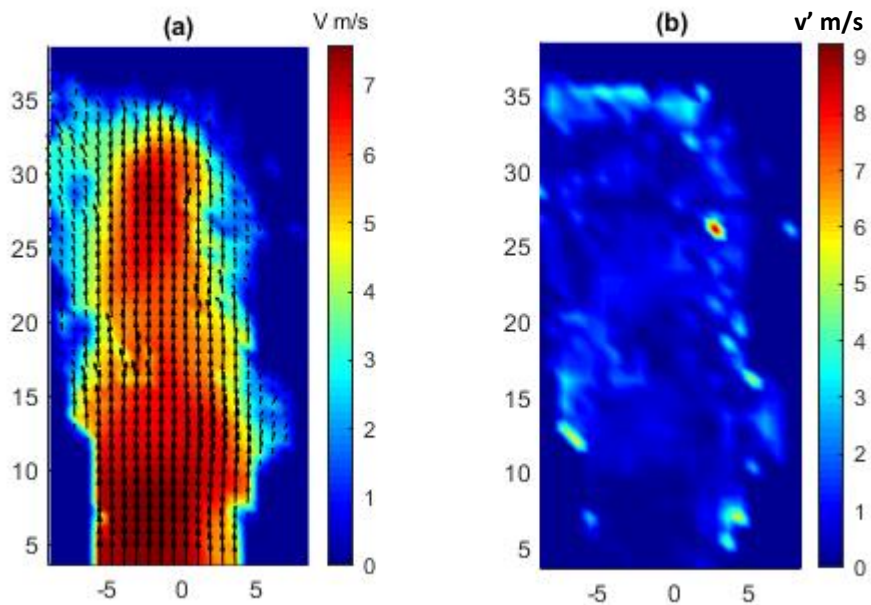
13 Figure 4.12b and Figure 4.13b shows the respective axial turbulence intensities for
14 the laminar and turbulence flows. As expected, the potential core areas were
15 characterized by lower turbulence intensities compared to the outer mixing
16 region. In the laminar case (Figure 4.16), there is nearly a constant low turbulence
17 intensity in core area which persists at different heights considered. In contrast,
18 the turbulence flow experienced higher turbulence (5 -20%) which increased
19 downstream as shown in Figure 4.17.

20



1

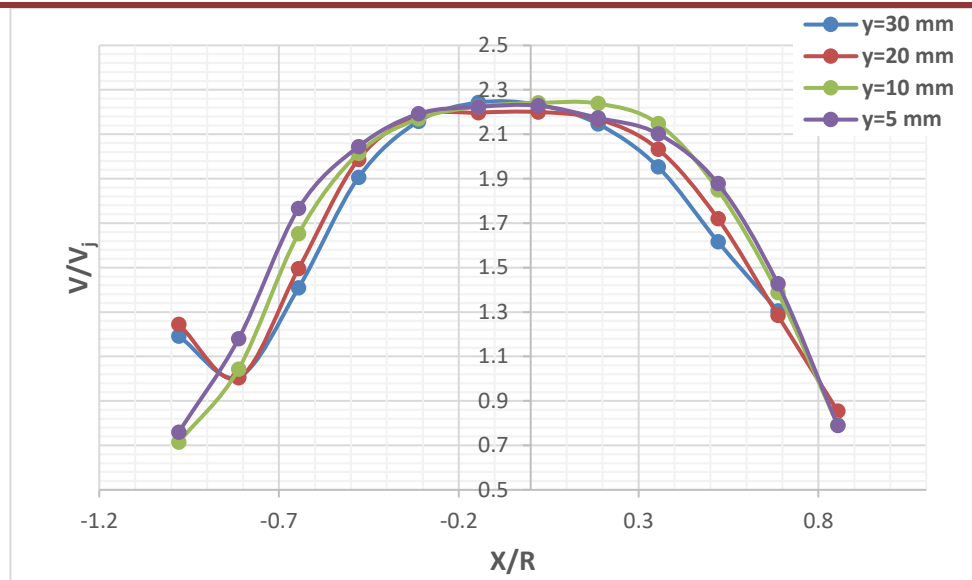
2 **Figure 4.12: Mean flow characteristic of the Laminar jet. (a) Mean velocity b) Mean**
 3 **axial turbulence intensity**



4

5 **Figure 4.13: Mean flow characteristic of turbulent jet. (a) Mean velocity b) Mean**
 6 **axial turbulence intensity**

7

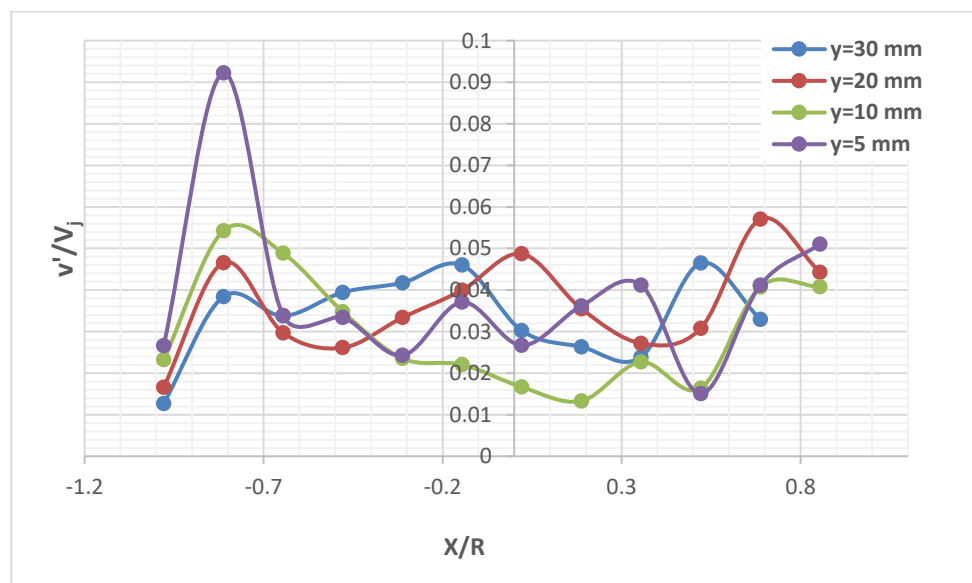


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2

Figure 4.14: Mean Axial Velocity profiles (Laminar flow, $V_j=1.15$ m/s, $R=5$ mm)

3

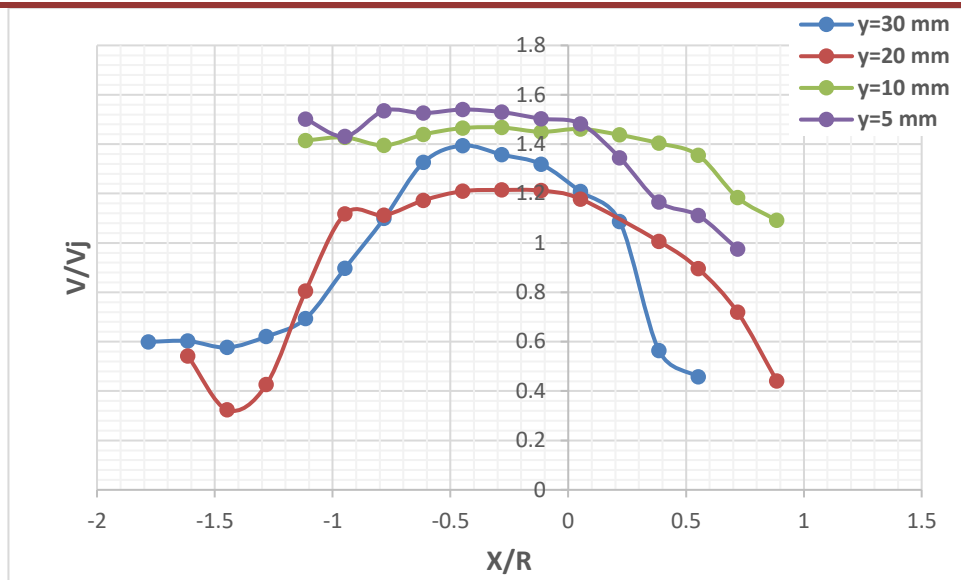


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Figure 4.15: Axial Turbulence Intensity profiles (Laminar, $V_j=1.15$ m/s, $R=5$ mm)

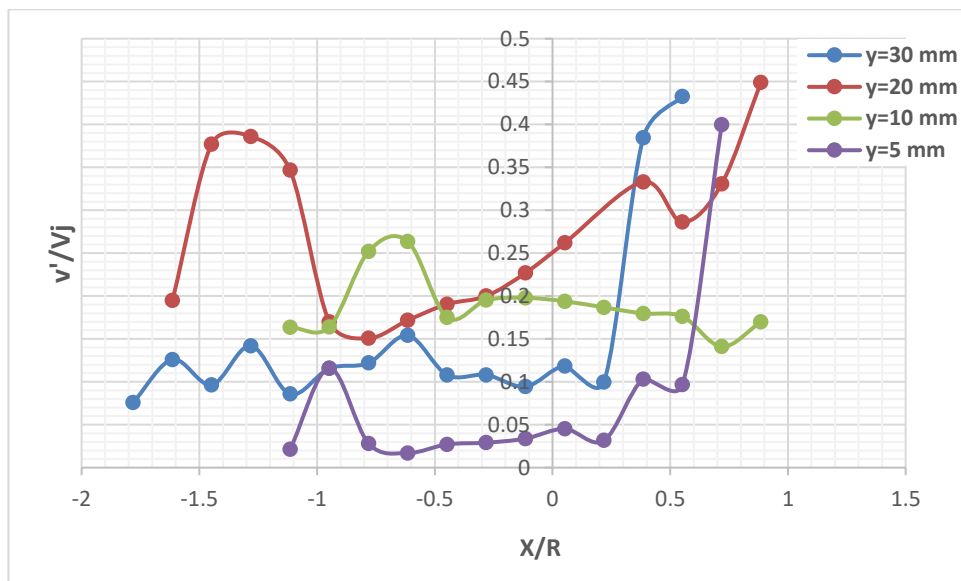
6



1

2 **Figure 4.16: Mean Axial Velocity profiles (Turbulent flow, $V_j=4.89$ m/s, $R=5$ mm)**

3



4

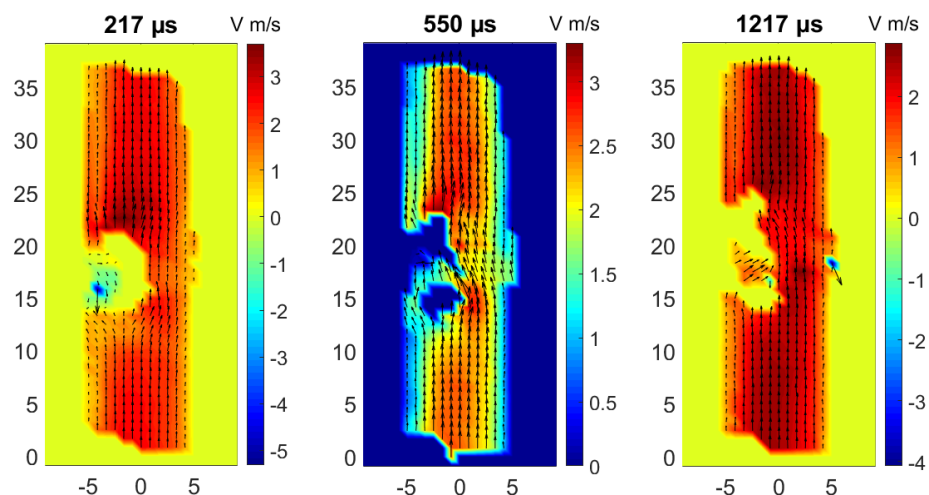
5 **Figure 4.17: Axial Turbulence Intensity profiles (Turbulent flow, $V_j=4.89$ m/s,**
6 **$R=5$ mm)**7 **4.4.2 Characteristic of the reacting flow**

8 Figure 4.18 is the instantaneous velocity vector fields of the burner flow at three
 9 different stages of the flame kernel development (217, 550 and 1217 μ s) for the
 10 laminar case. The first main observation in the first frame (at 217 μ s) is the zero-
 11 velocity region near the kernel trailing edge due to rotation effect of the vortex

1 generated as the front lobe moves towards the laser. This vortex motion is also
2 responsible for the swirling flow observed due to mixing of the upstream and
3 downstream flow.

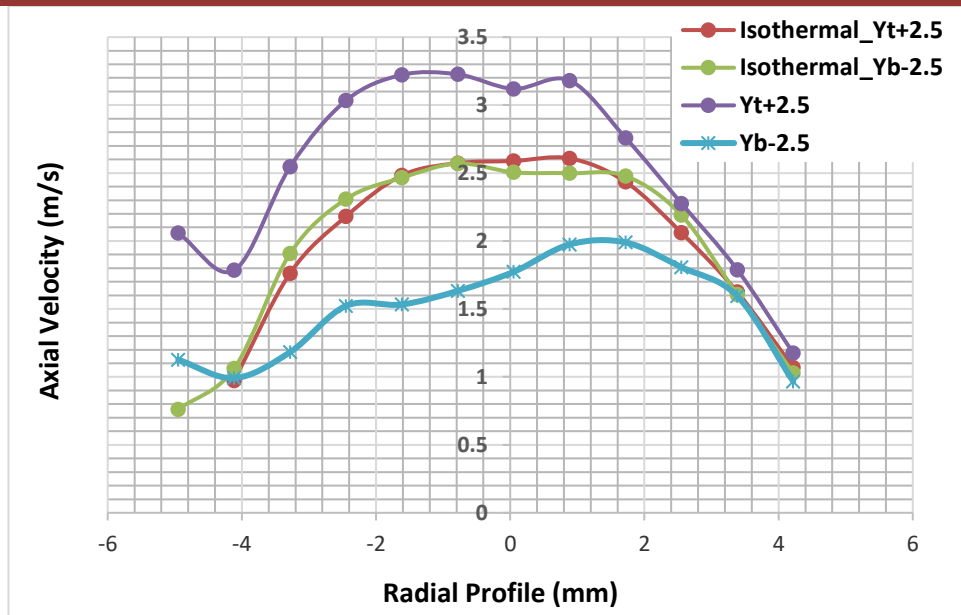
4 The second observation is the significant difference in the velocity field of the
5 upstream and downstream flow. As observed in the three frames, a conspicuous
6 peak in axial velocity (about 3 m/s) is noticed in the downstream flow just ahead
7 of the flame front. This peak is caused by the acceleration of the flow due to rapid
8 thermal expansion of the flame kernel and had been observed previously in a fully
9 developed flame [156]. Additionally, the velocity vectors downstream appear
10 vertical as in the isothermal laminar flow (before ignition). In contrast, the axial
11 velocity of the upstream flow just behind the flame front is significantly reduced
12 especially in the first two frames. The differences in behaviour of the both the
13 upstream and downstream flow is further illustrated by axial profile plots in
14 Figure 4.19. By further investigation of the flow field changes with development of
15 the flame kernel, it was discovered the initial jump in downstream peak velocity
16 decreased with time as the kernel grew in size until it approaches the isothermal
17 flow field velocity (see Figure 4.20). Likewise, the initial drop in upstream velocity
18 increased with as the kernel grew developed until it attains the isothermal flow
19 field velocity (see Figure 4.21)

20



21

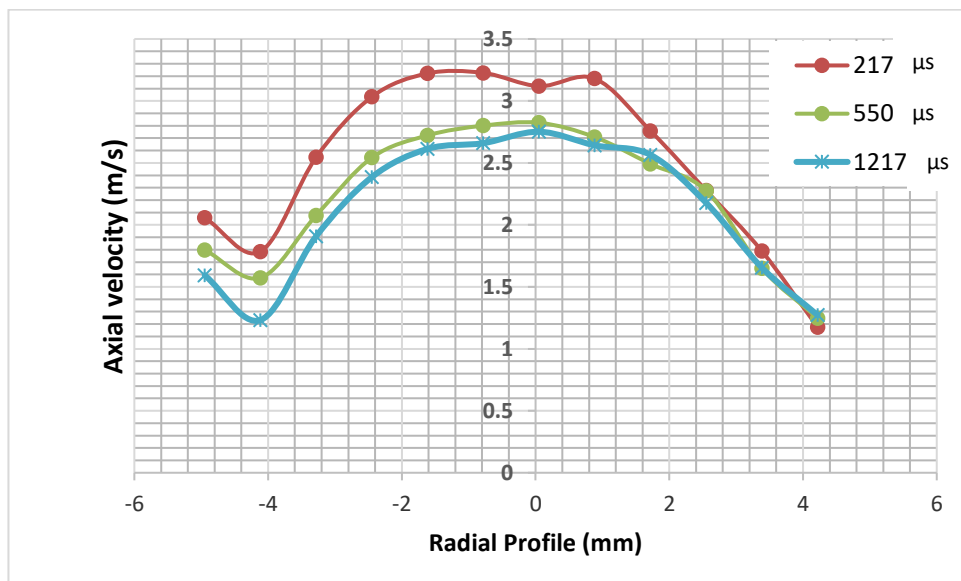
22 **Figure 4.18: Instantaneous Velocity Vector plots in Laminar Reacting flow**



1

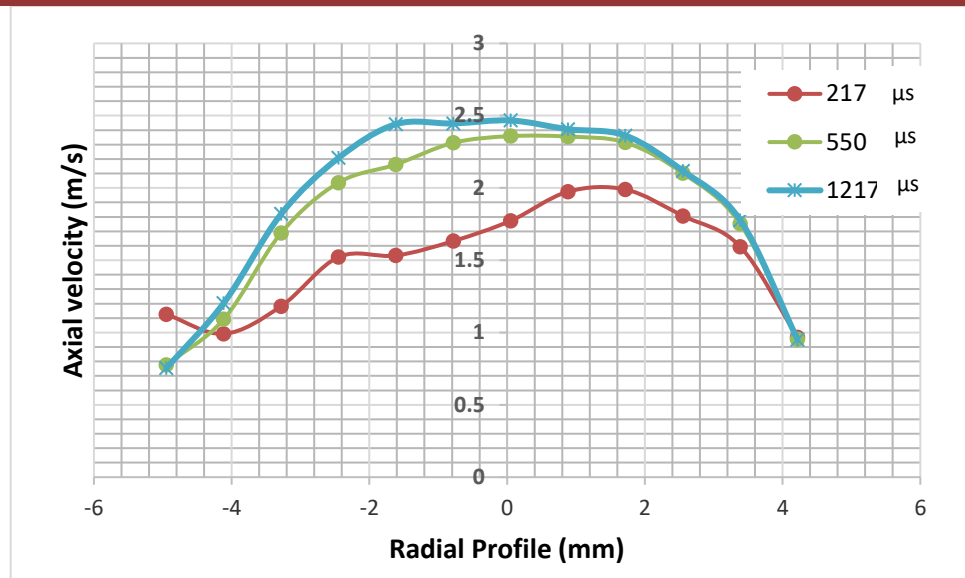
2 **Figure 4.19: Profiles plot of Axial Velocity at 217 μ s delay compared with the**
 3 **Isothermal flow. [Yb,Yt] - Upstream and Downstream of Flame front**

4



5

6 **Figure 4.20: Radial profiles of Axial Velocity at location (Yt+2) mm downstream**



1

2

Figure 4.21: Radial profiles of Axial Velocity at location (Yb-2) mm upstream

3

4.5 A note about the results

4

It is important to mention that there were gaps in data set owing to limitations. An obvious discrepancy is the lack of symmetry in the radial profile of the axial flow velocity vectors. This resulted from calibration errors. As earlier explained in section 3.51, setting up the High-speed PIV system required a target object of known dimensions which was captured before the actual experiment commenced. Using this target capture dimensions, subsequent images were scaled to determine the x-, y- dimensions in both pixels and mm. The calibration of the flow field also involved setting up the central axis of the flow which ideally should be along the centre of the burner diameter. Therefore, the use of a target object for this exercise instead of an actual flow no doubt created the discrepancy in the measured data.

14

Another limitation encountered is the fact that only single capture preliminary results were used for this investigation. It was originally planned to acquire a series of cycle ignition measurement which will be compared and average to obtain a true picture of the flame kernel development. However, this was not possible due to equipment breakdown. After months of waiting, the equipment was repaired but then moved to other projects which made it difficult to conclude the experiment coupled with time limitation. It is therefore advised that the results reported in this chapter be used with utmost caution.

21

1 4.6 Conclusions

2 In this chapter, results of the instantaneous flame kernel characteristics and its
3 surrounding flow velocity field were reported based on instantaneous high-speed
4 images acquired during laser-induced ignition of stoichiometric Methane/Air
5 mixture. A summary of the main findings is given below:

- 6 • The flame kernel visualization study using laser sheet tomography provided
7 insight on the changes in the geometric features of the flame kernel during
8 its development. At ignition, an elliptically shaped plasma of hot gas was
9 first observed. This then developed into a two-lobe toroidal structure
10 (which is symmetric about the laser axis) by 50 μs . The next feature that
11 appeared before 167 μs was a front lobe resulting in an asymmetric toroidal
12 flame kernel. The images also reveal how the front lobe results from the
13 motion of a centreline jet flow generated by large contra-rotating upstream
14 and downstream vortices.
- 15 • Variations between the measured initial plasma radius (at about 1 μs) and
16 the Blast wave theory showed some differences with respect to the flow.
17 For example, in laminar flow case, R was 13.9% less while in the turbulent
18 flow case R was 4% more.
- 19 • The displacement locations of the upstream and downstream flame fronts
20 (i.e., Y_+ , Y_-) differed depending on the flow condition. For example, in the
21 laminar flow mixture, the kernel flame front locations showed a growth
22 curve nearly symmetric about the laser axis, while for the turbulent case,
23 this symmetry is lost due to displacement of the kernel centroid. In
24 addition, the spatial evolution of the kernel height (Y) shows that in the
25 laminar flow, Y reaches the burner diameter (i.e., 10 mm) in about 0.7 ms
26 while in the turbulent flow, it took only 0.3 ms to reach this height.
- 27 • Changes in axial growth rate of the kernel show there were three key
28 development stages. From an initial shock speed 23 times the unstretched
29 laminar value (S_L), the kernel decayed in stages to a stable speed close to S_L
30 in 1 ms. A reasonable deduction from this is that the initial growth was
31 dominated by induced gas velocity of the shock waves, the second phase of
32 growth was the result of both the induced gas velocity and chemical

1 reaction while the final phase was predominantly due to reaction
2 chemistry.

- 3 • The velocity field visualization obtained by PIV analysis revealed key
4 information on the structure of the flow field in both reacting and non-
5 reacting state. Prior to ignition, the axial velocity plots reveal an
6 axisymmetric flow structure with maximum velocity in the potential core
7 area which decreased radially to a minimum near the flow edges. In the
8 laminar flow, the axial velocity profiles did not change significantly with
9 height, however in the turbulent flow the velocity profiles decreased with
10 increase in height. Unlike the flow velocity, the turbulence intensity was
11 stronger in the mixing region (i.e., near the flow edges) than in the potential
12 core (i.e., centre of the flow). Moving downstream, the general trend reveals
13 an increasing intensity, already this was somewhat irregular in the
14 turbulent flow.
- 15 • Following ignition, the observed flow field velocities were different from
16 the isothermal case. Due to the rapid thermal expansion of the flame, an
17 increased axial velocity peak is observed in the downstream region just
18 ahead of the flame front. Upstream the flow profile shows change in near
19 the flame front which caused a skew in the symmetry of the developing
20 flame kernel. In contrast, a reduced axial velocity peak is observed in the
21 upstream region just behind the flame front. Further investigation of the
22 radial profiles of the axial velocity revealed that this variation in velocity
23 peak existed only within certain distances (below 2.5 mm) from the flame
24 fronts.

1 Chapter 5 OH* chemiluminescence characteristics of the 2 Flame Kernel and its dependence on physical parameters.

3 5.1 Introduction

4 This chapter presents further characterisation of the laser-ignited flame kernel
5 through analysis of the mean OH* chemiluminescence images and its dependence
6 on two physical parameters: pulse energy and flow velocity. The study is divided
7 into three main sections. The first section begins with qualitative characterisation
8 of the flame kernel based on the mean chemiluminescence images obtained at
9 various ignition delays. Further edge extraction of the mean kernel images enabled
10 quantitative description of the kernel characteristics by the flame front location
11 and its propagation rate. In addition, the relative changes in OH* concentration
12 with time was described from the integrated and maximum OH* intensity. While
13 kernel location is useful for predicting the burning rate, the relative OH*
14 concentration gives information about the heat release rate due to chemical
15 reaction. The second included investigation is the effect of pulse energy on the
16 evolution of the flame kernel structure for the flow condition. Finally, the flow
17 velocity effect on the kernel properties were investigated in section three.

18 5.2 Test conditions

19 Table 5.1 gives a summary of the test conditions for the three investigations
20 conducted in this chapter. As shown in the table, the initial run (test 1) was
21 conducted with laminar methane-air mixtures ($\phi = 1.0, U_0 = 1.15 \text{ m/s}$) to gain a
22 general overview of the flame kernel development characteristics. This was
23 followed by a study of the pulse energy effects (test 2) and finally a study of flow

1 velocity effects (test 3) was conducted. For all the tests, methane-air mixtures of
 2 constant stoichiometric composition were used.

3

4 **Table 5.1: Summary of test conditions**

Test	1	2	3
Flow parameters:			
Equivalence Ratio, ϕ	1.0	1.0	1.0
Air Flow Rate (litres/min)	23.80	23.80	23.80, 42.86, 61.90, 71.43
CH ₄ Flow Rate (litres/min)	2.5	2.5	2.5, 4.5, 6.5, 7.5
Bulk flow velocity, U_0 (m/s)	1.15	1.15	1.15, 2.08, 3.00, 3.46
Flow Reynolds Number, Re (Pa. s)	1694	1694	1694, 3050, 4405, 5083
Adiabatic flame temperature, T_{ad} (K)	2226	2226	2226
Laminar flame velocity, S_L (cm/s)	41	41	41
Nitrogen co-flow (litre/min)	3	3	3
Laser parameters:			
Pulse energy, E (mJ)	32	32, 64, 96, 128	32
Laser frequency, f (Hz)	2	2	2
Laser wavelength, λ (nm)	1064	1064	1064

5

1 **5.3 General OH* chemiluminescence characteristics of the flame kernel**

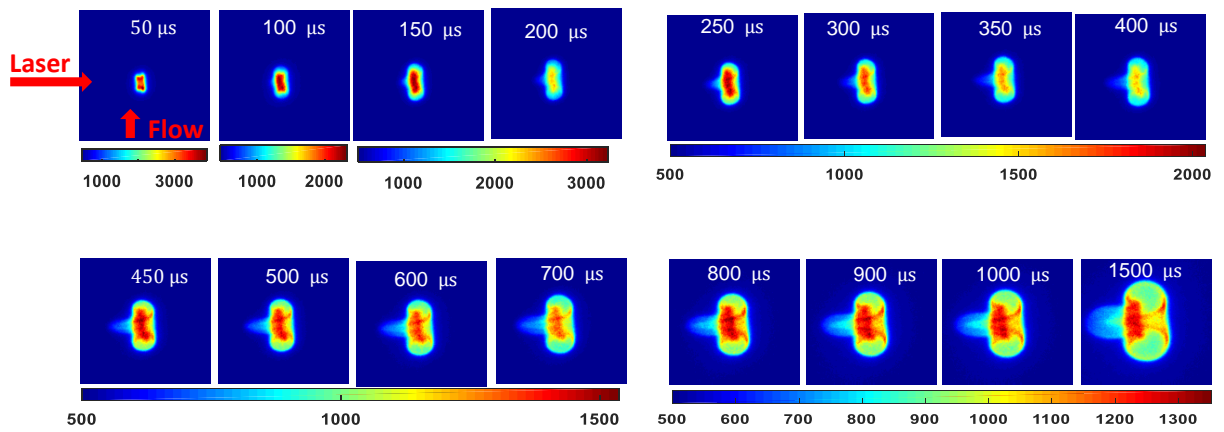
2 **5.3.1 Visual characteristics of the Flame Kernel**

3 Before studying the parametric dependence, it was necessary to first characterise
4 the flame kernel based on the mean OH* chemiluminescence images acquired for a
5 single test condition [CH_4/Air ($\emptyset = 1.0, U_0 = 1.15 \frac{\text{m}}{\text{s}}$), $E_p=32 \text{ mJ}$].

6 The first observation from the acquired images is the changes in kernel shape as it
7 develops from a two-lobe symmetric toroid to a three-lobe asymmetric toroid.
8 Figure 5.1 shows the sequence of mean OH* chemiluminescence images acquired
9 between 50 μs and 1.5 ms time delays with respect to the ignition pulse. The first
10 kernel image observed at 50 μs is a butterfly-shaped (or two-lobe) toroidal
11 structure propagating symmetrically above and below the laser axis. Previous
12 studies observed similar shape for both laser ignition and electric spark ignition
13 [92],[100]. The origin of this feature has been attributed to an induced inward flow
14 resulting from an over-expansion of the shock wave, emanating outward from the
15 region of the discharge at a very high velocity [138]. From 100 μs onwards, a third
16 lobe (a.k.a. front lobe) begins to emerge from the edge of the kernel facing the
17 incident laser beam thereby leading to an asymmetric structure. Previous studies
18 [92],[100] has shown that this third lobe is a unique feature of laser ignition and it
19 propagates towards the direction of the incident laser. Hence, for the first 1.5 ms,
20 the kernel shape is predominantly a three-lobe toroid with rapid growth along the
21 central axes of the lobes. Figure 5.2 shows that beyond 1.5 ms, local quenching
22 occurs at the front lobe while growth is continuous along the symmetric (i.e.,
23 upper, and lower) lobe axes. The resulting final shape of the kernel is similar to
24 that of a typical flame ball propagating mostly in the upstream and downstream
25 directions of the flow. In this study, the flame is prevented from stabilizing to allow
26 repeated measurement. Therefore, the flame front growth downstream is
27 enhanced by the flow while the flame front growth upstream is restrained by the
28 flow. In addition, propagation of the flame in both directions is marked by
29 changing radius of curvature.

1 The second observation from the images is the progressive change in reaction
2 intensity and distribution of reaction zones within the kernel as it evolves.
3 Between 50 μs and 100 μs , the peak intensity of OH* dropped from 3800 AU to
4 2300 AU due to cooling of the hot gases which make up the initial kernel. At 150 μs ,
5 a new peak in OH* intensity occurs which is the first indication of the kernel
6 survival. Subsequent kernels showed a continuous decay in peak OH* until a
7 minimum peak is attained beyond which the peak rises again due formation of a
8 self-sustained flame. Further observation the image colourmap shows that the
9 distribution of the OH* within the kernel differs at different stages of development.
10 At the initial stages (50 - 300 μs), the OH* is evenly distributed, hence the kernel is
11 composed of an isotropic reaction zone. A possible explanation for this is that
12 initial kernel growth is predominantly due to the presence of hot gases which
13 originated from the initial hot plasma. At the mid stage of development (450 -
14 1500 μs), the distribution of OH* reveals the kernel is composed of anisotropic
15 reaction zones with peak intensity within the middle region. A possible
16 explanation for this is that kernel growth during this period is the result of both
17 induced gas flows and chemical reactions. This could also be the result of mixing of
18 the reaction zone with the flow due to vortex induced motion which may lead to
19 multiple flame fronts within the kernel. As shown in Figure 5.2, the colourmap of
20 the late kernels (3 - 10 ms) shows that the reaction zones (i.e., flame fronts) were
21 concentrated at the edges. Unlike the early kernels, the inner region of low
22 intensity may indicate the presence of burnt gases. These observations also give an
23 idea of the two-way interaction involving the flame-flow mixture and flame-burnt
24 products.

25



1

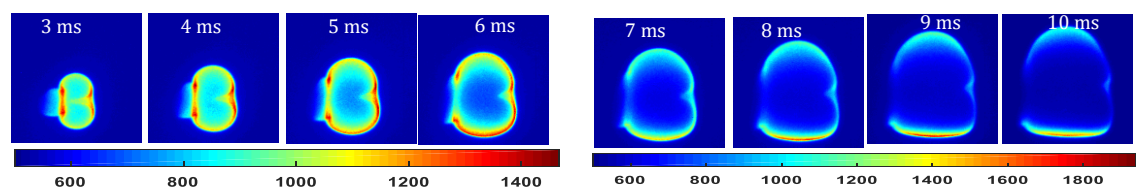
2

Figure 5.1: Mean OH* chemiluminescence images showing 50 – 1500 μs Kernel Development (Image FOV: 28x28 mm²)

3

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6

Figure 5.2: Mean OH* chemiluminescence images showing 3 – 10 ms Kernel evolution (Image FOV: 56x56 mm²)

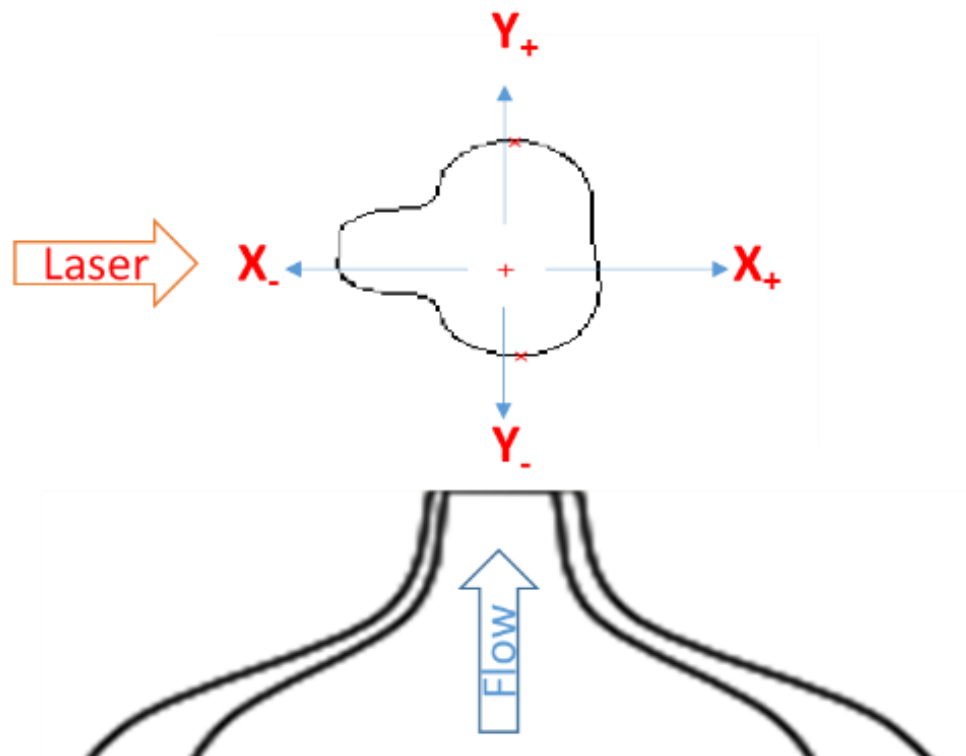
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9 5.3.2 Analysis of the Flame Kernel Geometric Characteristics

10 From the extracted edges of the mean OH* chemiluminescence images, geometric
 11 analyses were performed to ascertain the growth of the flame kernel with time.
 12 The first characteristics investigated is the size and its flame front location in both
 13 vertical axis (i.e., flow axis) and horizontal (i.e., laser axis). Figure 5.3 is a
 14 schematic of the measurement plane showing the locations of the flame front
 15 relative to the spark location. Y_+Y_- represents the flow axis while X_+X_- represents
 16 the laser axis.

17



1
2 **Figure 5.3: Measurement plane showing the flow and laser axes of the Flame**
3 **Kernel**

4
5 Figure 5.4 shows the displacement of the flame front tip from the ignition point
6 downstream and upstream the flow axis. Following ignition, the displacements of
7 the upstream and downstream flame fronts reveal a gradually decaying growth up
8 to 0.8 ms when the growth become steady. From this point the downstream flame
9 front maintain a steady linear growth. Likewise, the upstream flame front
10 maintained a steady linear growth until it reached its maximum displacement of
11 12 mm in about 7 ms. The combined growth of the flame fronts (i.e., Y_+ and Y_-)
12 reveals a nearly symmetric growth about the laser axis. This is however not the
13 true situation as revealed by the two-dimensional scatter plot of axial flame tip
14 displacement (Figure 5.5).

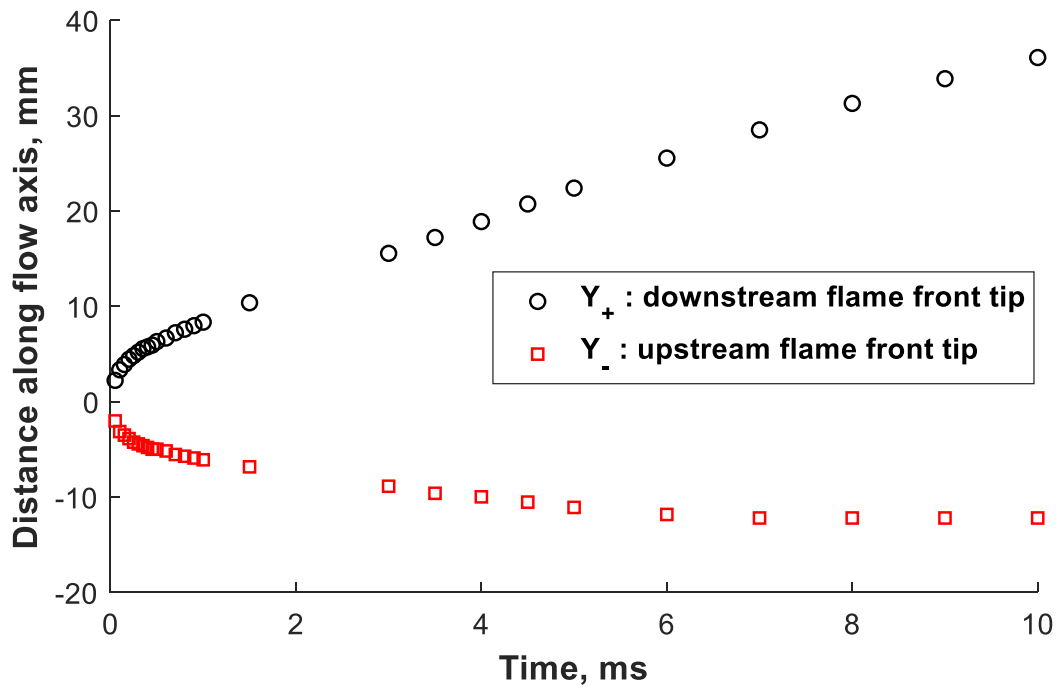
15 Figure 5.6 shows the displacement of the flame front tip from the ignition point
16 along the laser axis. Unlike the axial flame fronts, the transverse displacement of
17 the fronts (i.e. X_+ and X_-) is clearly non-symmetric. This is expected due to the

1 presence of the front lobe. Following ignition, the displacements of the flame fronts
2 along opposite directions of the laser axis is characterised by an almost linear
3 growth with sudden change in slope at specific instances. For example, the
4 displacement of the flame front away from the laser was marked by sudden decline
5 at 0.1 ms, thereafter the growth was almost linear with very little decay. Kernel
6 was very rapid during the first 1.5 ms. Likewise, displacement towards the laser
7 was relatively steady until 1.5 ms, when an abrupt change in slope occurred and
8 then a steady linear growth onwards. The abrupt change in the slope of X_{-} at 1.5
9 ms occurred because of local quenching due to high stretching of the front lobe.

10 To further compare growth along both axes, the dimensions of the kernel were
11 evaluated from the differences in kernel tip displacement. Figure 5.7 compares the
12 evolution of the kernel height (H) with its width (W). According to the figure, only
13 small differences were observed in the first 1.5 ms between the kernel dimensions
14 with H slightly greater than W. The small difference in the early development is
15 due to the presence of the front. In past studies [97], this has been attributed to the
16 asymmetry in the dimensions of the initial spark kernel. This knowledge may be
17 useful when deciding the best orientation of the incident laser beam for a
18 particular combustion system. Beyond 1.5 ms, the difference between H and W
19 became increasingly wider due to flow velocity effects. In the current setup, it
20 takes about 2 ms for the kernel to reach the spatial size of the burner (i.e., diameter
21 =22 mm) along the flow axis while it takes about 2.5 ms to reach the same
22 dimension along the laser axis.

23

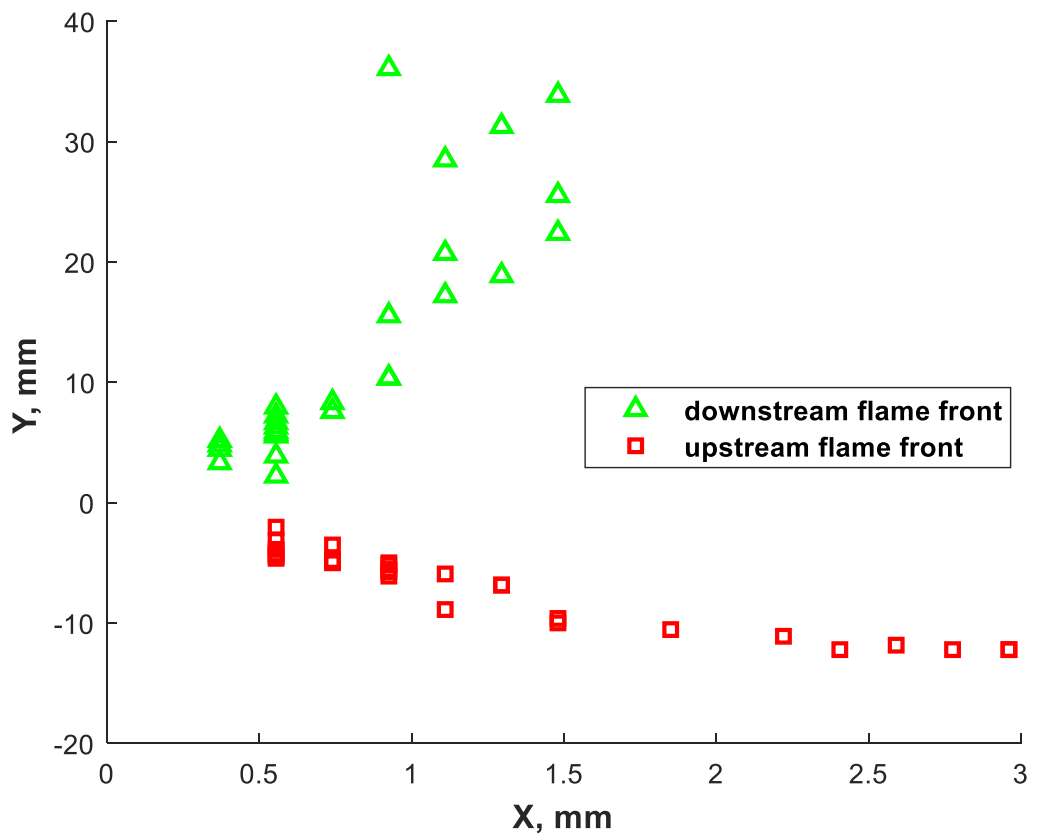
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Figure 5.4: Displacement of the flame front tip along flow axis

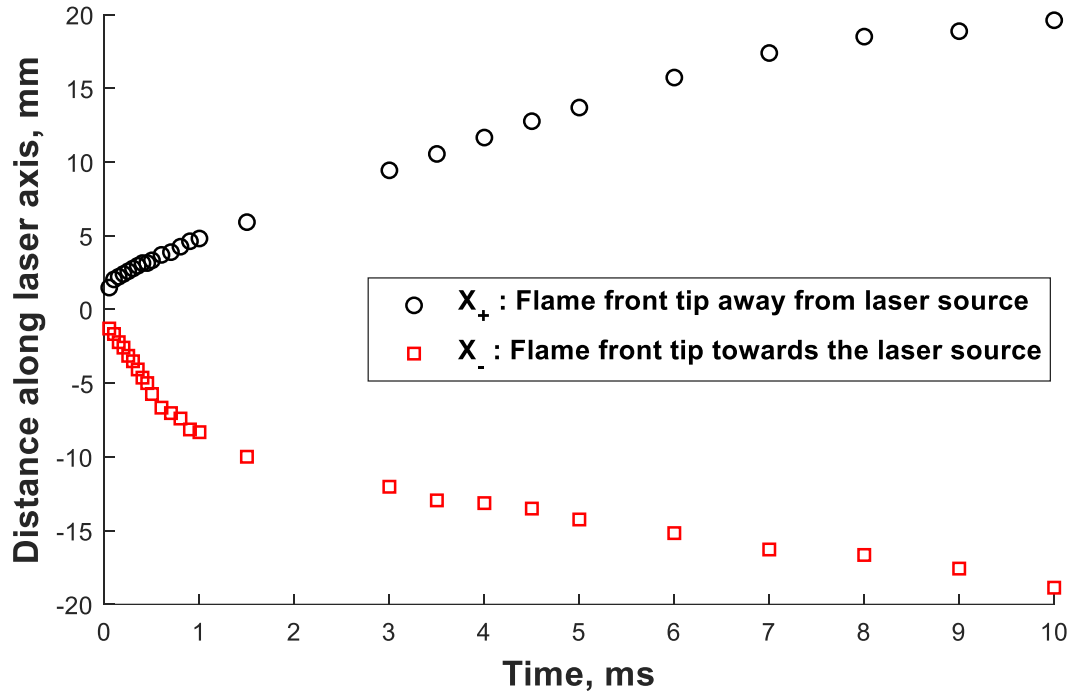


3

4

Figure 5.5: Two-dimensional scatter plot of axial flame front tip

1

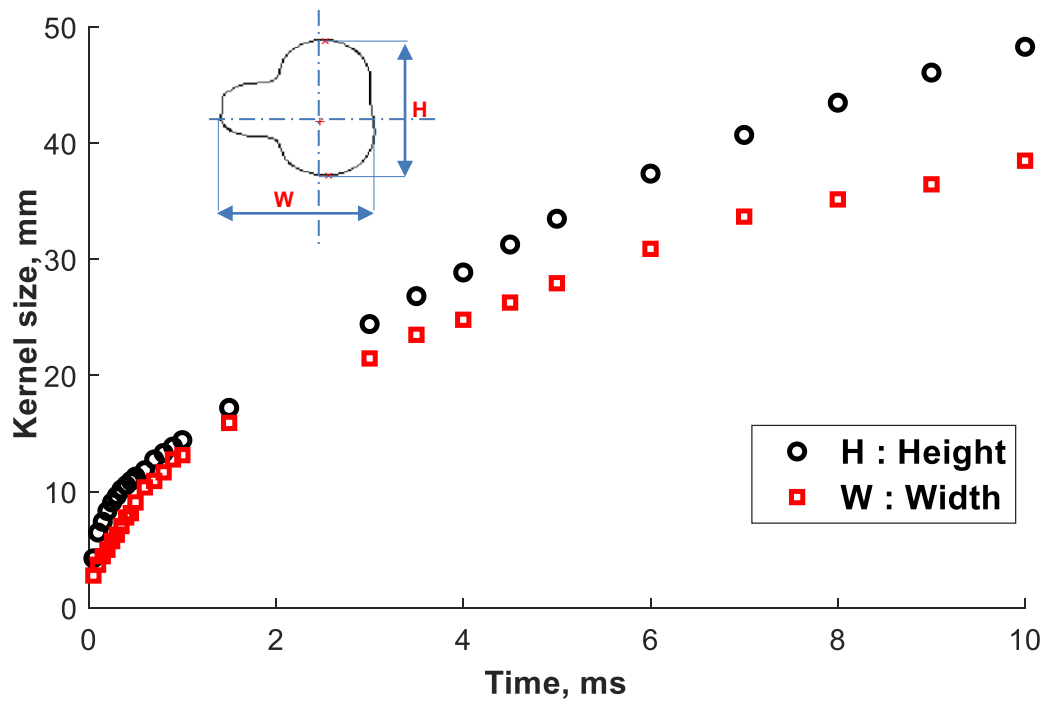


2

3

Figure 5.6: Displacement of the flame front tip along Laser axis

4



5

6

Figure 5.7: Evolution of the Flame Kernel dimensions

1 Next, an analysis of the growth rates of flame front tips was performed from the
2 measured tip displacements. To determine the kernel growth rates, the time
3 derivatives of the tip displacement were obtained according to the following
4 equations:

5
$$S_{y+} = \frac{dy_+}{dt} \quad (5.1)$$

6
$$S_{y-} = \frac{dy_-}{dt} \quad (5.2)$$

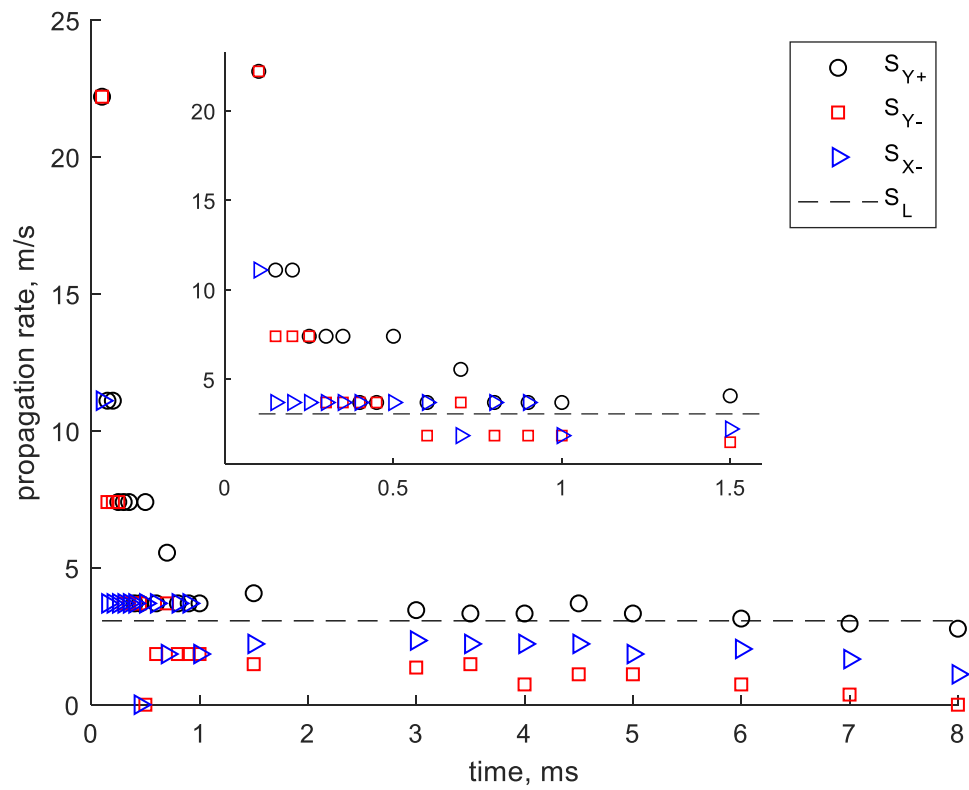
7
$$S_{x-} = \frac{dx_-}{dt} \quad (5.3)$$

8

9 Figure 5.8 is a graph showing the kernel propagation rates along the three main
10 directions of propagation. From the graph, the general trend is that the growth
11 rates decay from a highly stretched value to an asymptotic value close to the
12 unstretched laminar flame growth rate (S_L). Where S_L is evaluated from the
13 product of the unstretched laminar flame velocity (u_L) and the expansion
14 coefficient which is the ratio of fresh gas density to the density of the burnt
15 product (i.e. $S_L = \rho_u/\rho_b * u_L$). Analysis of the growth rate of each flame front
16 highlight the existence of three key development stages. For example, S_{y+} dropped
17 from an initial high value of 22 m/s to a steady value of 11 m/s in the first 0.15 ms.
18 This was the first phase of growth dominated by induced gas velocity of the shock
19 waves. Between 0.2 and 0.25 ms, S_{y+} decayed from 11 m/s to a second stable
20 speed of about 7.5 m/s which marked the formation of an igniting kernel. This
21 marked the beginning of the second stage of development when the kernel
22 propagates due to combined influence of chemical reactions and induced gas
23 velocity. The third and final major decay in growth rate occurred between 0.5 and
24 0.8 ms when S_{y+} dropped to a steady velocity of 3.5 m/s. Beyond this point, the
25 downstream flame front maintained a stable velocity which fluctuated around the
26 mean value of 3.3 m/s when growth is mainly due to chemical reactions.
27 Therefore, it is needful to investigate the sensitivity of the flame front growth to
28 these competing factors under changing test conditions.

1 Figure 5.9 compares the actual speed of the flame front along the three main
2 directions of propagation. To accurately compare them, a velocity compensation is
3 applied by considering the bulk flow velocity (U_0) which affects growth along the
4 flow axis. Therefore, a value of 1.15 m/s is added S_{y-} to while the same value is
5 subtracted from S_{y+} . Such compensation is based on the assumption that the
6 horizontal component of flow velocity along the flow axis is negligible, hence, it
7 will only be valid under a laminar flow condition. The first observation highlighted
8 by this figure is that the growth rate of each flame front decay from a highly
9 stretched value to the same asymptotic value below the unstretched laminar flame
10 growth rate (S_L). The second observation is that there is variation in stretch
11 between the flame fronts during the early development stage. For example, at 0.1
12 ms, following the appearance of the front lobe, S_{y+} was 18 m/s higher than S_L
13 while S_{x-} is only 8 m/s higher. At 0.15 ms, the variation reduced to 6.9 m/s for S_{y+}
14 0.64 for S_{x-} . At the late kernel stage, the same value of growth rate is observed
15 along the different directions of propagation.

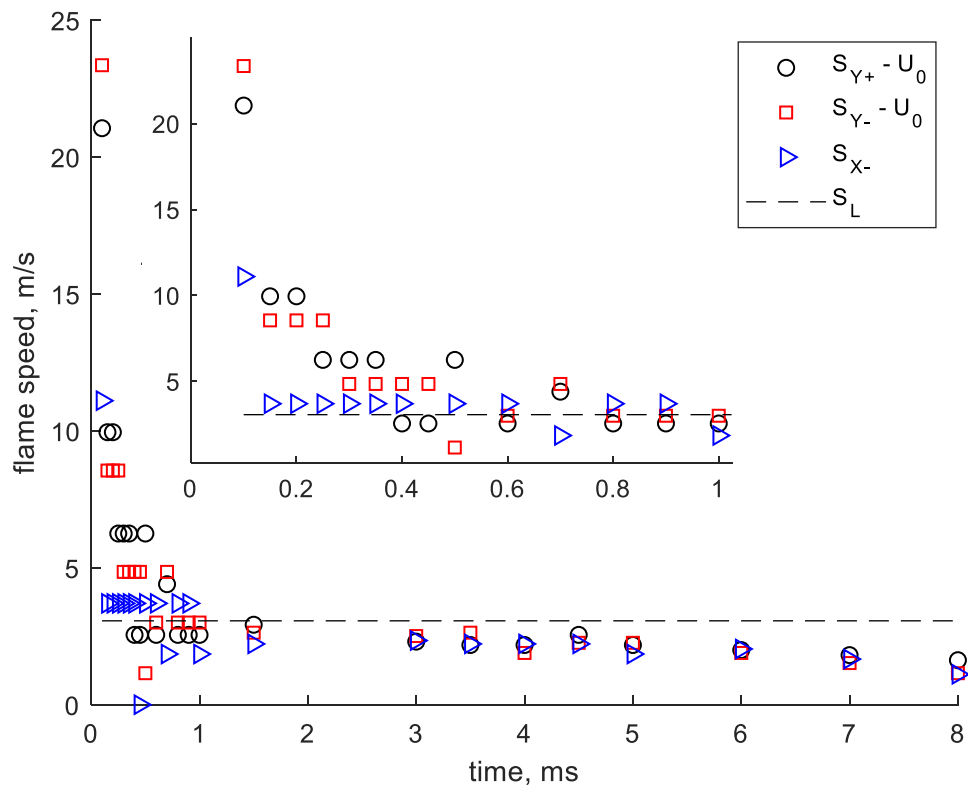
16



1

2 **Figure 5.8: Propagation rate along different directions of the flame front.**

3



1

2 **Figure 5.9: Growth rate of the flame front along the three-lobe directions**

3

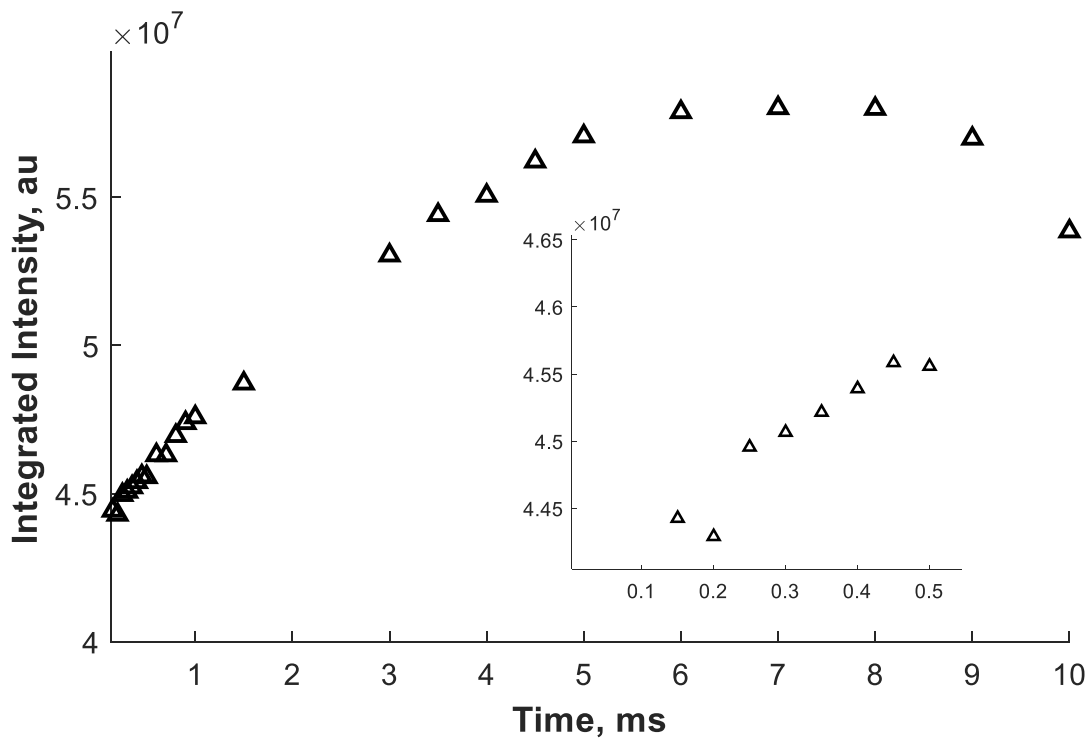
4 **5.3.3 Analysis of the Flame Kernel OH* Intensity**

5 Having studied the dynamics of the flame kernel geometry, it was necessary to
 6 investigate the dynamics of the OH* intensity which could provide an idea of the
 7 coupling between the fluid dynamics and chemical kinetics of the flame kernel.
 8 From the raw images, a quantitative analysis was also performed to show the
 9 general trend in OH* intensity characteristics of the flame kernels. For a reliable
 10 measure of the OH* intensity evolution, the images analysed were captured from
 11 the same field of view (351x351 pixels) and with an intensifier gain setting of 200.

12 Figure 5.10 is a plot of the spatially integrated OH* intensity from 150 μ s to 10 ms.
 13 The evolution of the spatially integrated OH* intensity provided information about
 14 the strength of the flame kernel as well as further evidence of the key transition

1 phases of kernel development. The measurement shows that the integrated OH*
2 intensity decreased in value between 0.15 to 0.2 ms. This initial decrease indicates
3 transition from a quenching flame kernel to a more reactive flame kernel. The
4 event could also be interpreted as the chemical induction phase. Beyond this point,
5 the integrated OH* intensity rises continuously in a non-linear fashion. A rising
6 trend in the integrated OH* intensity indicates a developing kernel with a higher
7 chance of survival because the cumulative heat release was increasing with kernel
8 expansion. Between 6 ms and 8 ms, a plateau is observed in the integrated OH*
9 intensity indicating a fully developed flame. Beyond 8 ms, a downward trend is
10 observed in the integrated OH* intensity which may be due to heat losses resulting
11 from interaction of the flame kernel with the Nitrogen used as blow-off gas. By
12 relatively comparing the integrated OH* intensity with the kernel dimensions, it
13 can be deduced that both predicts the key transition points of the early
14 development such as the end of chemical induction and the time when growth is
15 purely due to chemical reaction.

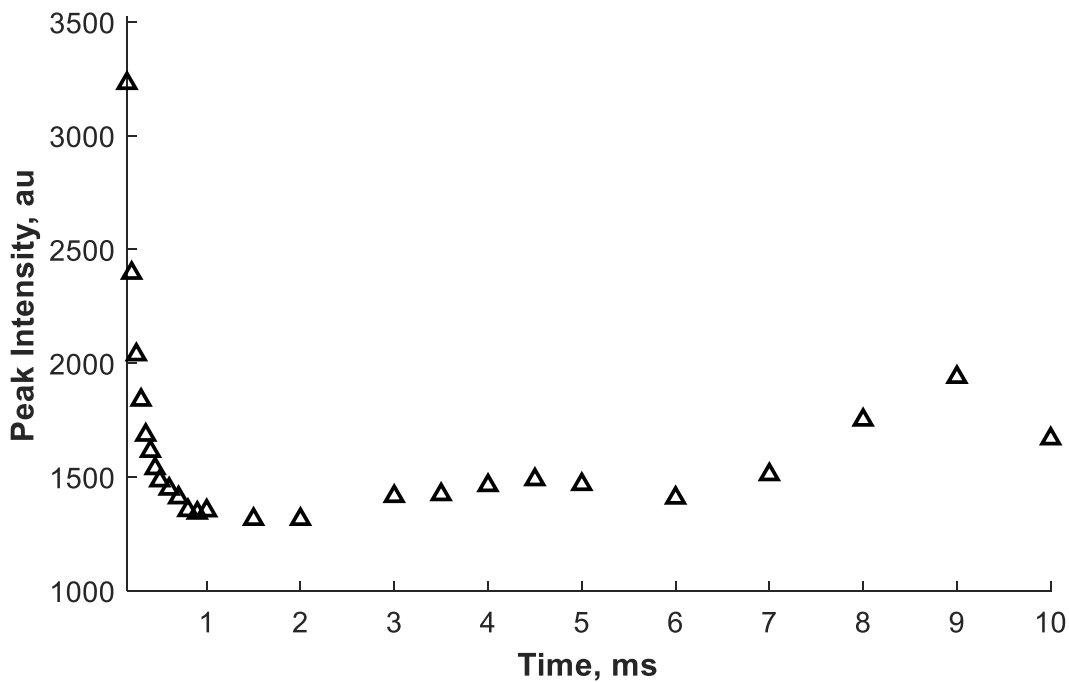
16 Figure 5.11 is the evolution of peak OH* intensity from 150 μ s to 10 ms time delay.
17 The peak intensity indicates the measure of heat release due to the balance
18 between radical production rate and consumption rate. Just like the cooling curve
19 of most gases, the observed trend in peak in OH* intensity shows a continuous
20 decay until it reaches a minimum value 1314 AU around 1.5 ms. After reaching this
21 minimum value, a slow recovery is observed in the peak OH* intensity reaching a
22 trough value of 1500 AU at 4.5 ms and a second trough value of 2000 AU at 9
23 ms. At the developed stage, the kernel's ability to maintain intensity values above
24 the minimum peak is necessary to prevent quenching of the flame kernel and
25 indicates continuous heat release due to chemical reaction. Also, correlation can be
26 made between the time kernel reaches its minimum peak OH* intensity and the
27 period when the propagation rate of the flame fronts reaches an asymptotic value.



1

2 **Figure 5.10: Evolution of the mean integrated OH* intensity from 150 μ s to 10 ms**

3



4

5 **Figure 5.11: Evolution of the mean peak OH* intensity from 150 μ s to 10 ms**

1

2 **5.4 Effects of Laser Pulse Energy**

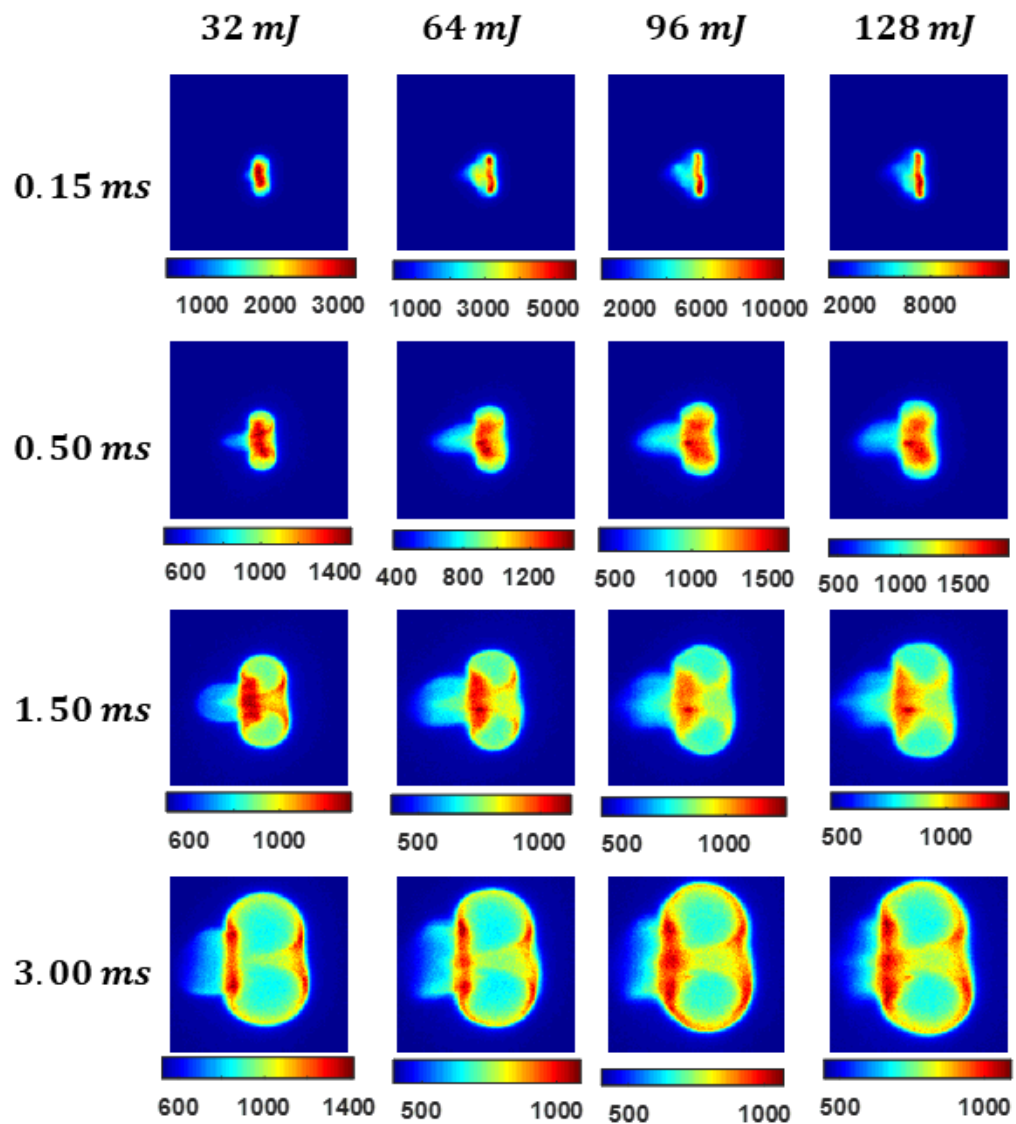
3 Having studied the fundamental characteristics relevant to flame kernel
4 propagation, the next stage of the study was to investigate the effect of the key
5 control parameters. In this section, the effect of laser pulse energy on the flame
6 kernel characteristics was investigated using four variations of pulse energy (i.e.,
7 32, 64, 96 and 128 mJ). For fair comparison, a constant flow condition (same as
8 that used in Test case 1) is maintained throughout.

9 Figure 5.12 visualizes different development stages of the flame kernel (150 μ s,
10 500 μ s, 1500 μ s and 3000 μ s) due to changing pulse energy. As in earlier
11 observation, the flame kernel shapes were identical for all cases each evolving
12 from a two-lobe symmetric toroid to a three-lobe structure with the late kernels
13 developing into a flame ball shape. Overall, with a higher pulse energy, the upper
14 and lower toroid develops faster producing a bigger sized kernel at a particular
15 time delay from the beginning of an ignition event. This increase in kernel size is
16 attributable to higher plasma induced gas velocity associated with higher pulse
17 energy leading to higher stretching of the kernels. In the late kernels, the impact of
18 increased pulse energy is not so significant compared with the early kernels,
19 especially for higher pulse energies. A second visible effect of higher pulse energy
20 on the early kernels is that the front lobe development is faster for higher energies
21 and their lifespan is shorter due to higher stretch rates. This is due to the
22 stretching effects of high energy pulses which can lead to premature extinction of
23 the kernel. Based on the images captured at 150 μ s delay, overstretching resulted
24 in separation of the front lobe flame front from the rest of the kernel at higher
25 pulse energies. A third visible effect of pulse energy can be observed from the color
26 scale of images acquired at the same time delay. For example, at 150 μ s each
27 increase in pulse energy produced a corresponding increase in peak OH* intensity.
28 By 1.5 ms, each increase in pulse energy had no effect on the peak OH* intensity. In

1 contrast to the early kernel, a downward trend was observed in peak OH* intensity
2 due to increased energy at 3 ms.

3

4



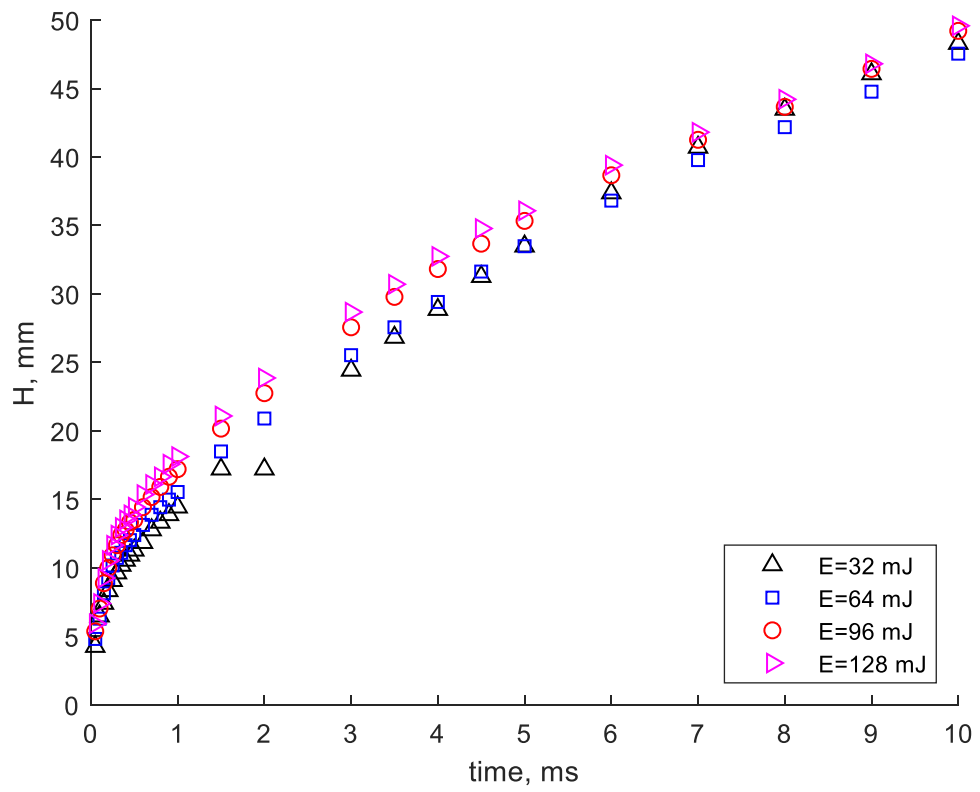
5

6 **Figure 5.12: Visual comparison of flame kernel development at different pulse**
7 **energies (FOV: 28x28 mm²)**

8

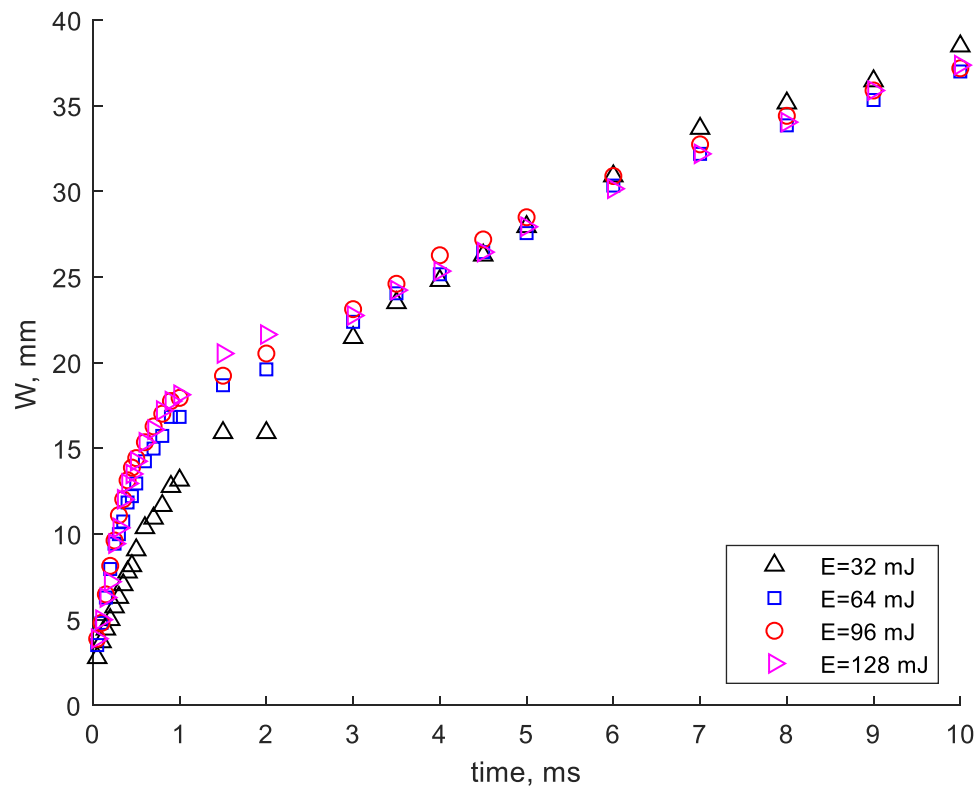
1 Figure 5.13 and Figure 5.14 shows the comparative dimensions of the developing
2 flame kernel due to changing pulse energies. Both plots reveal a noticeable
3 increase in kernel size due to higher energy especially at the early development
4 phase. It is worth noting that there was a significant increase in size (especially W)
5 when the incident energy increased from 32 mJ to 64 mJ, however, further
6 increase in energy had little or no effect on the kernel size. This shows the
7 existence of a threshold incident energy for optimal ignition performance for the
8 selected focusing lens and flow condition. The observation agrees with previous
9 authors that spark energy deposition has a limiting efficiency depending on the
10 lens focal length [90],[157]. The current observation showed that further energy
11 increase beyond the threshold value could lead to premature extinction instead of
12 enhancing flame propagation. Further analysis showed the most enhancement in
13 early kernel growth rate occurred in the first 400 μs along X - when the pulse
14 energy increased from 32 mJ to 64 mJ. This further confirms the existence of a
15 threshold energy above which an increase in pulse energy had no impact on
16 growth rate. No significant changes were observed in the late kernel stage with
17 increase in pulse energy.

18



1

2 **Figure 5.13: Evolution of kernel height compared at different pulse energies**



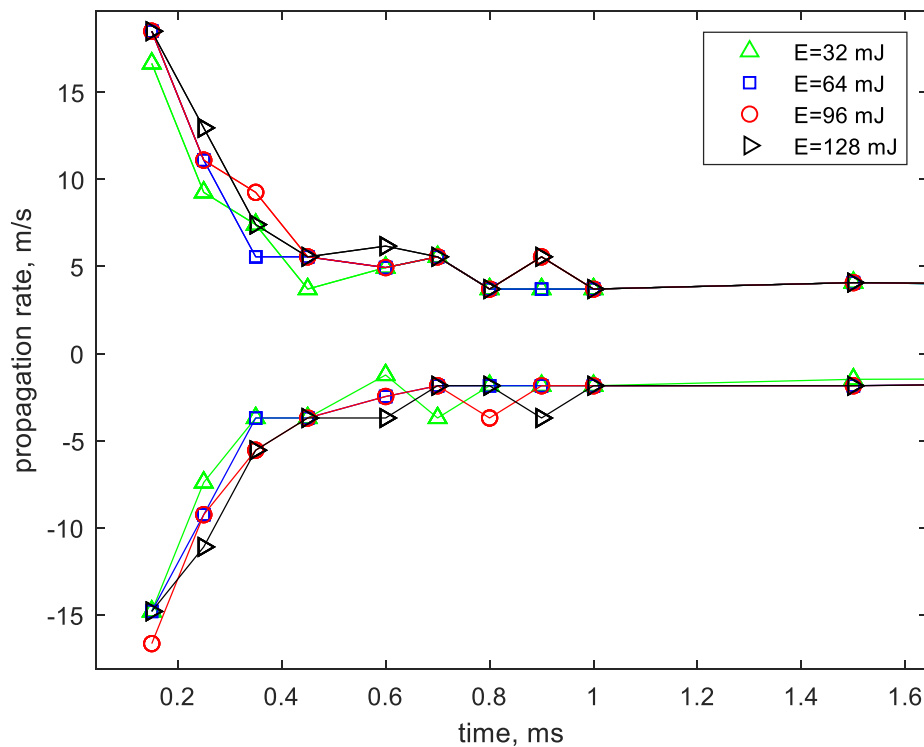
1

2 **Figure 5.14: Evolution of kernel width compared at different pulse energies**

3 Figure 5.15 compares the propagation rate of the flame front tip along opposite
 4 directions of the flow axis at varying pulse energies (i.e., 32, 64, 96 and 128 mJ). A
 5 common feature of the plots is that the growth is characterised by decay from a
 6 high initial velocity to a stable asymptotic velocity close to the laminar unstretched
 7 value. Overall, the kernel propagated faster with higher incident energy, but this
 8 happened before 1 ms, during the early kernel development. From 1 ms onwards,
 9 all the kernels propagated with the same steady state velocity which implies that
 10 stretch effect is only prevalent at the development stage.

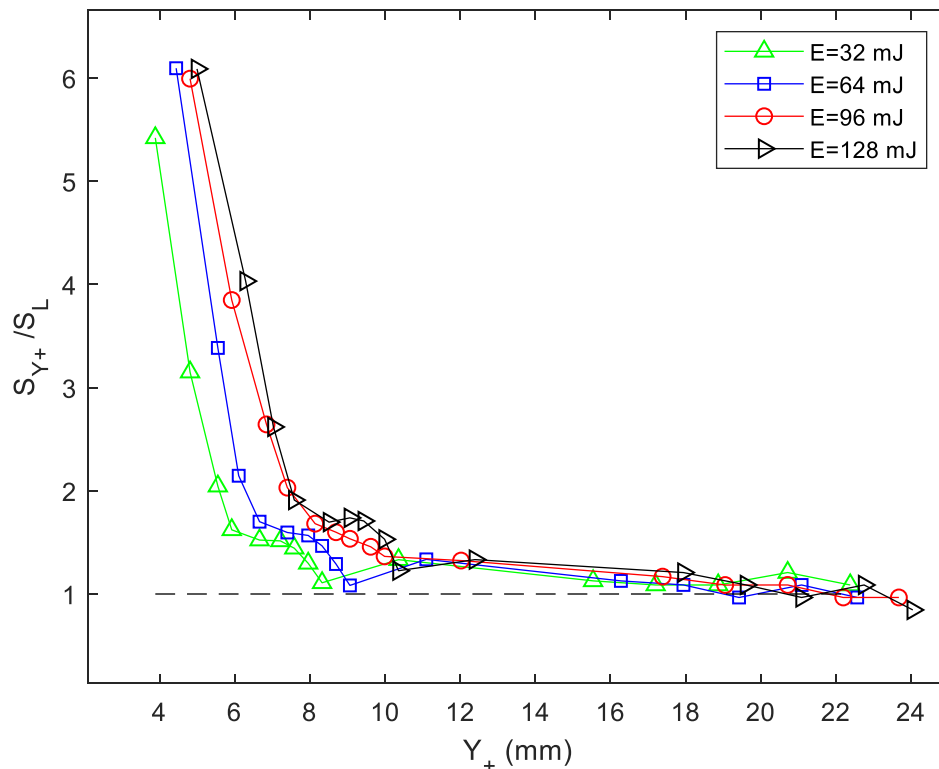
11 To better illustrate the relative changes in propagation rate and stretch effect of
 12 the incident energy, the normalised flame front propagation speed downstream is
 13 plotted as a function of the displacement as shown in Figure 5.16. As the figure
 14 shows, when the incident energy was 32 mJ, S_{y+} reached its asymptotic velocity
 15 after travelling 8.5 mm downstream which makes it the least stretched. On the
 16 other hand, when the incident energy was 128 mJ, S_{y+} reached its asymptotic

1 velocity after travelling 10.5 mm downstream which makes it the most stretched.
2 Furthermore, for the same displacement of 8 mm, the respective stretched speed
3 at 32, 62 and 128 mJ were 20, 20, 30, and 50% more than the upstretched laminar
4 value. The similar stretch between the two flame fronts at 96 and 128 mJ further
5 confirms the existence of a threshold incident energy above which further increase
6 has no impact on the growth. The results also shows that the incident energy has
7 no impact the growth of any of the flame kernels after extending 10.5 mm
8 downstream during which a constant velocity is maintained.



9

10 **Figure 5.15: Axial propagation rate at changing incident pulse energy.**



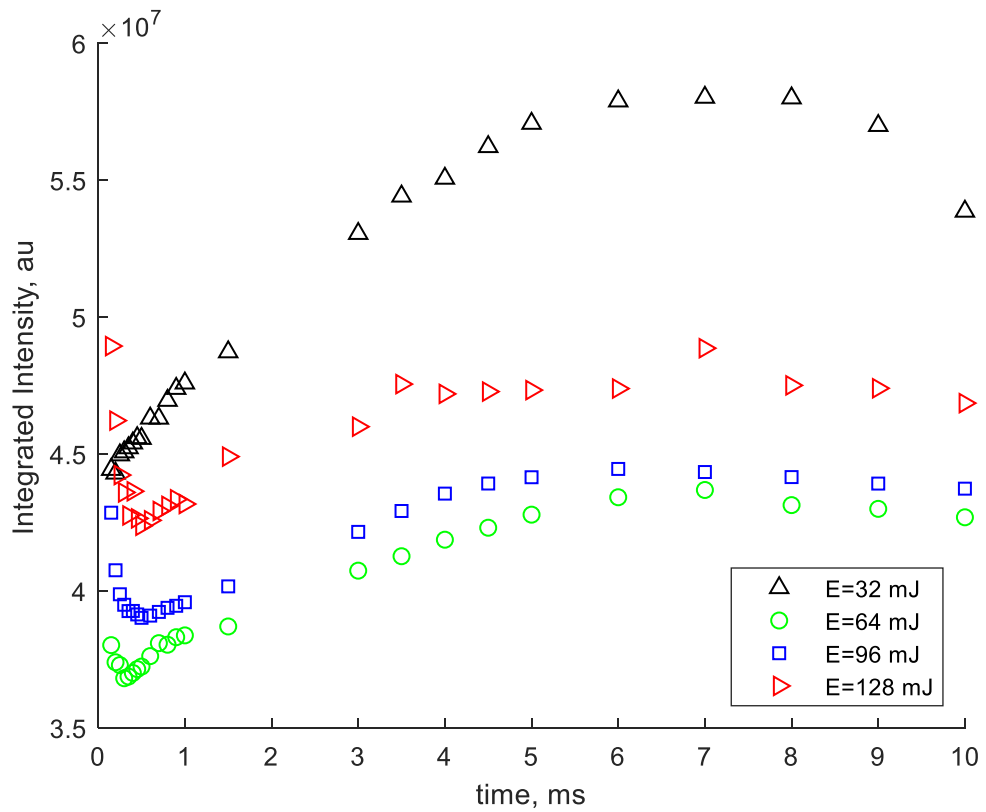
1

2 **Figure 5.16: Normalised propagation rate vs displacement at changing incident**
3 **pulse energy.**

4 Figure 5.17 shows the effect of increasing pulse energy on the integrated OH*
5 intensity. The first main observation from the results is that the value of the
6 integrated OH* intensity falls and then rises in a non-linear fashion and the time at
7 which the minimum integrated OH* intensity occurs is delayed with a higher pulse
8 energy. Therefore, it can be inferred that the higher the pulse energy, the longer
9 the chemical induction time. Following transition to an igniting kernel, the
10 cumulative heat release rises for each case which is marked by a continuous rise in
11 the integrated OH* intensity until a maximum value is reached. From the graphs, it
12 is evident the mean integrated OH* intensity is highly sensitive to increase in pulse
13 energy from 32 mJ to 64 mJ where a decrease in integrated OH* intensity was
14 observed. Further increase in energy from 64 mJ to 128 mJ resulted an increase in
15 the integrated OH* intensity but remained below the initial values at 32 mJ. This
16 shows that the total heat release was increasing with energy increase but not at
17 the optimum level.

1 An investigation of the variation in peak OH* intensity for different incident
2 energies is shown in Figure 5.18. From the graph, the initial values of the peak
3 intensity increased in accordance with the magnitude of pulse energy. However,
4 this trend did not last beyond 500 μ s, as the intensities decayed to the minimum
5 values and then a subsequent increase which marks a growing self-sustaining
6 flame. Following the formation of a self-sustaining flame, the peak OH* intensity
7 decreased with energy increase from 32 to 64 mJ. Further increase in energy
8 however did not produce any further change as observed in the kernel width.

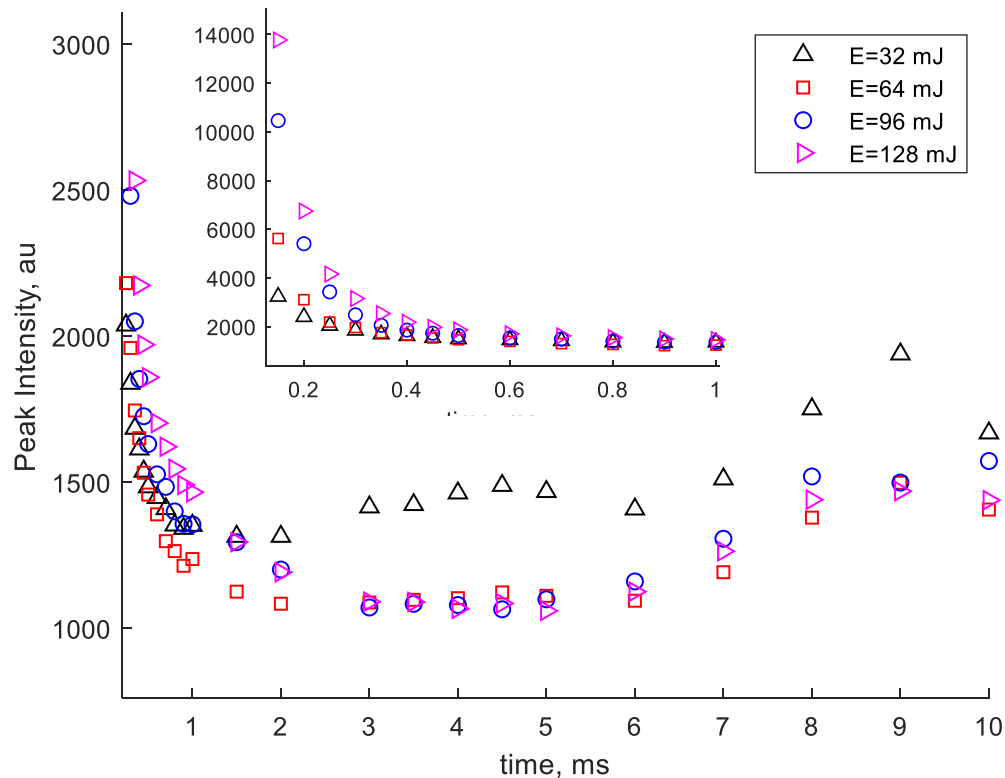
9 By considering all the observation, a conclusion can be drawn that the optimal
10 ignition energy should be decided based on the OH* production instead of the
11 flame stretch since the initial stretch resulting from higher pulse energies did not
12 produce bigger flame balls in the end. In the present investigation, a 32 mJ energy
13 would be ideal, however, it will be helpful to investigate if incidents energies below
14 32 mJ produced more OH*. It is logical that such investigation is done since the MIE
15 of the mixture is much lower (\sim 0.25 mJ).



1

2

Figure 5.17: Effect of pulse energy on the mean integrated OH* intensity



1

2

Figure 5.18: Effect of pulse energy on the mean peak OH* intensity

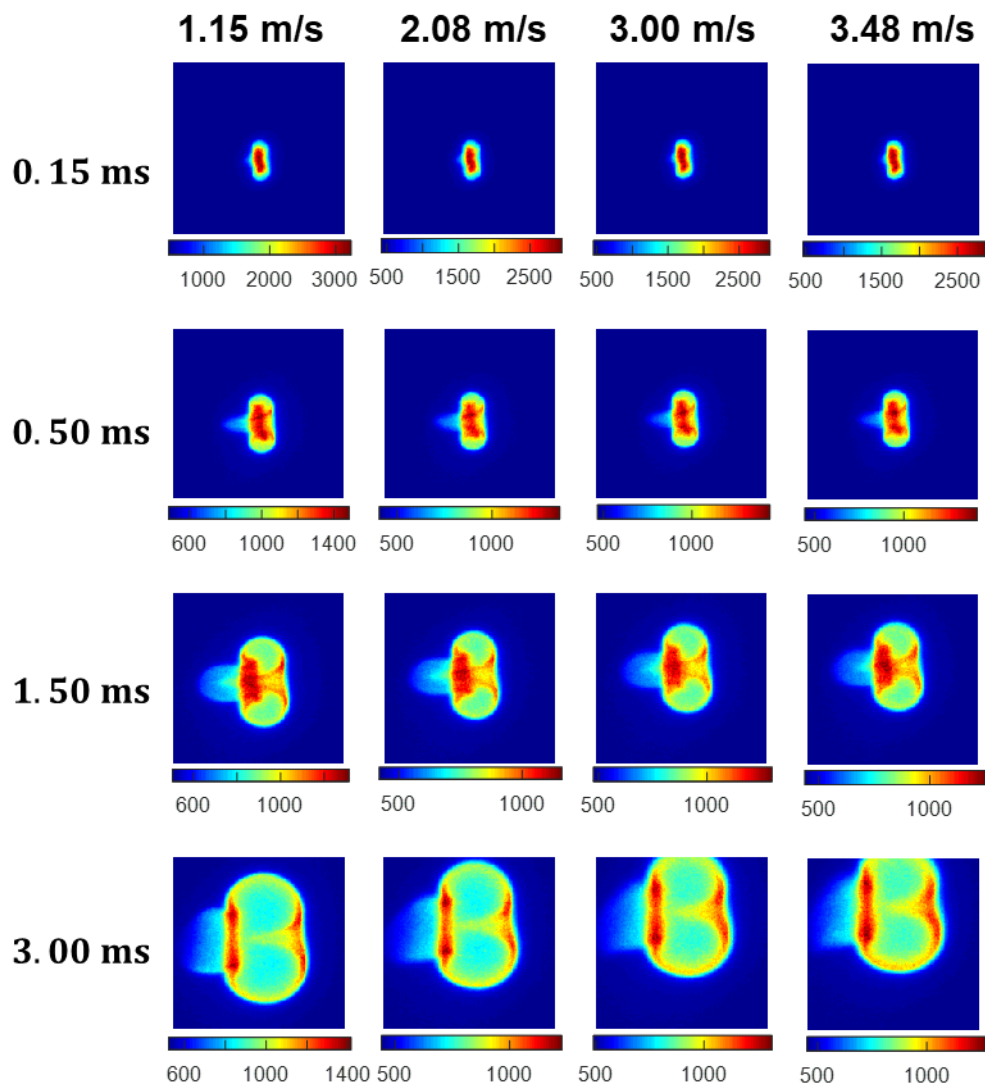
3

5.5 Effects of Flow Velocity

4 In this section, the effect of the gas flow velocity on the development of the flame
 5 kernel is reported. For fair comparison, experiments were conducted by
 6 maintaining a constant mixture composition and pulse energy (i.e., $\phi = 1.0$, $E =$
 7 32 mJ), while the flow velocity was varied in the following order: 1.15, 2.08, 3.0
 8 and 3.48 m/s.

9 Figure 5.19 visualizes different development stages of the flame kernel (150 μ s,
 10 500 μ s, 1500 μ s and 3000 μ s) due to variation in flow velocity. By comparing
 11 images acquired at the same ignition delays, no significant difference is observed
 12 in the kernel sizes and shapes especially at the development stage, however, the
 13 effect of increasing velocity is shown by the vertical displacement of the kernel
 14 centroid with respect to the spark position. At the developed stage, the pictures
 15 show that the kernel is under increasing amount of strain due to higher flow
 16 velocity. In addition, convective effect of the flow is shown by the fact that part of

1 kernel is no longer within the observation window. By comparing the image
2 colormap, it is evident that the surface of the flame front increases and becomes
3 less defined at higher flow velocities. This is of the wrinkling of the flame front due
4 to higher turbulence. In addition, there is hardly a noticeable change in peak OH*
5 intensity at different flow velocities but this would be analysed further.



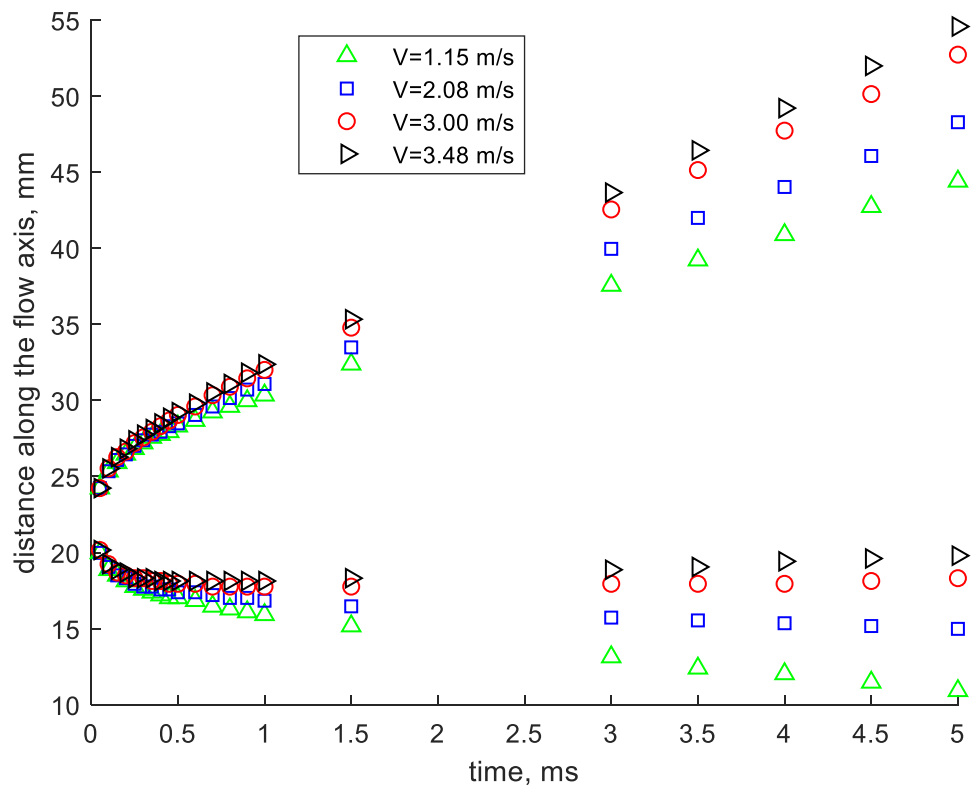
6

7 **Figure 5.19: Visualisation of flow velocity effects on kernel development**

8

1 To quantify the effect of increasing flow velocity on the kernel development and
2 propagation, each kernel characteristics were extracted and shown graphically on
3 the same timescale.

4 Figure 5.20 is a graphical picture of how the flow velocity influenced the
5 displacement of the flame front tip along the axis of the flow. The figure clearly
6 shows that the dominant effect of higher flow velocity is an increased displacement
7 of the flame kernel centroid which in effect resulted in an increased growth
8 downstream and decreased growth upstream. Further investigation showed that
9 difference between the upper and lower flame front tip for different flow velocities
10 were insignificant, hence, higher flow velocity had very little or no effect of the
11 kernel height. Another effect of high flow velocity is that of the increased strain on
12 the lower flame front. A clearer picture of the axial motion of the FF tips is shown
13 by the 2D scatter plot in Figure 5.21 which shows the displacement and strain
14 effect of increasing flow velocity on the flame kernel. Due to increasing strain
15 resulting from a higher flow velocity, further growth of the flame front upstream is
16 prevented resulting in a slightly wider kernels at higher flow velocities as shown
17 by Figure 5.22.

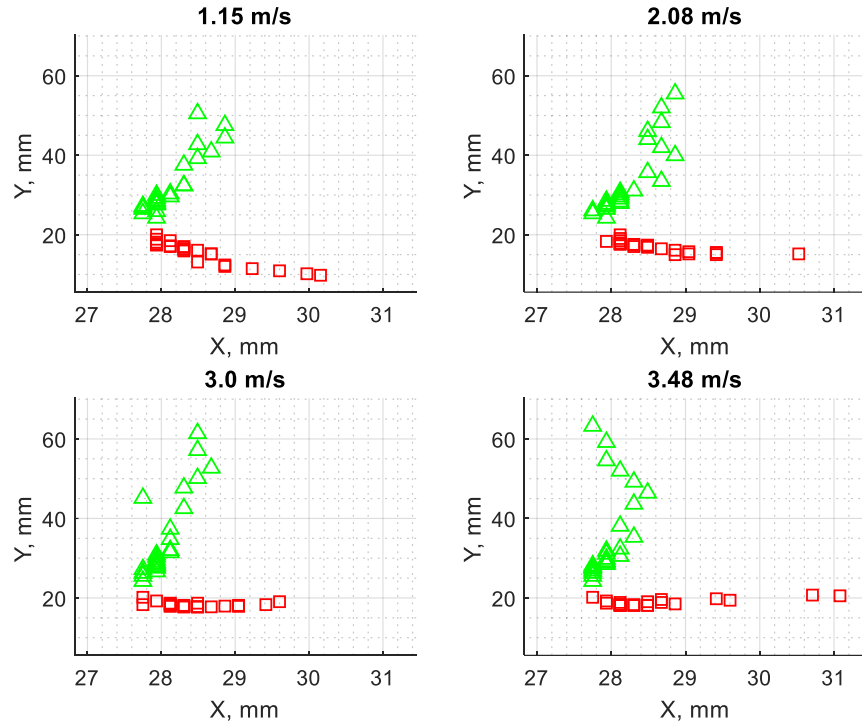


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2

Figure 5.20: Axial FF tip locations at varying Flow velocity

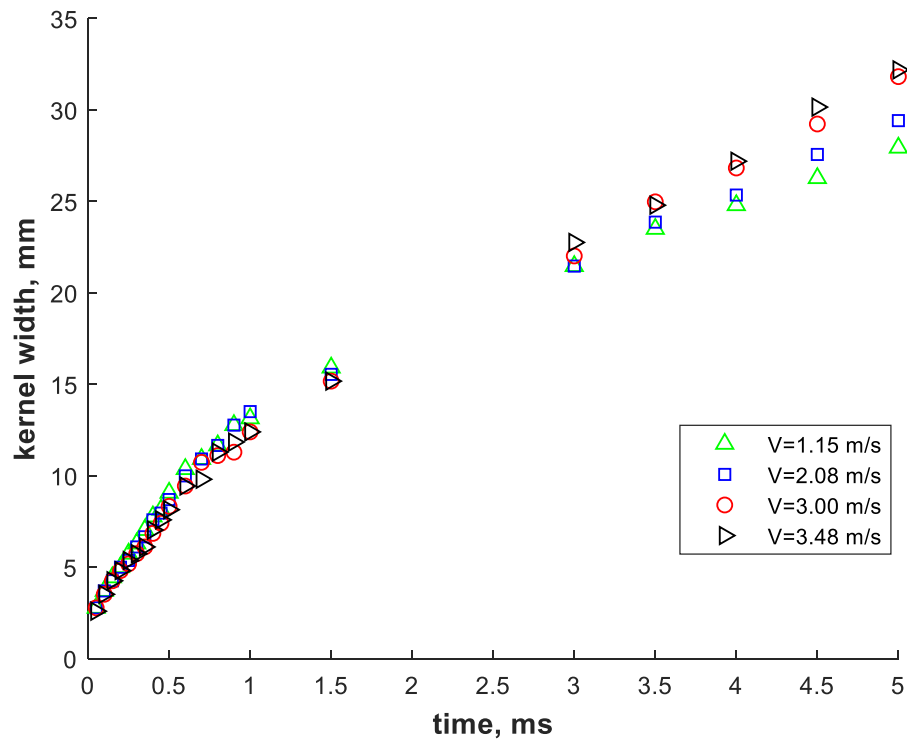
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Figure 5.21: Scatter plot of Axial tip location for varying velocity of flow



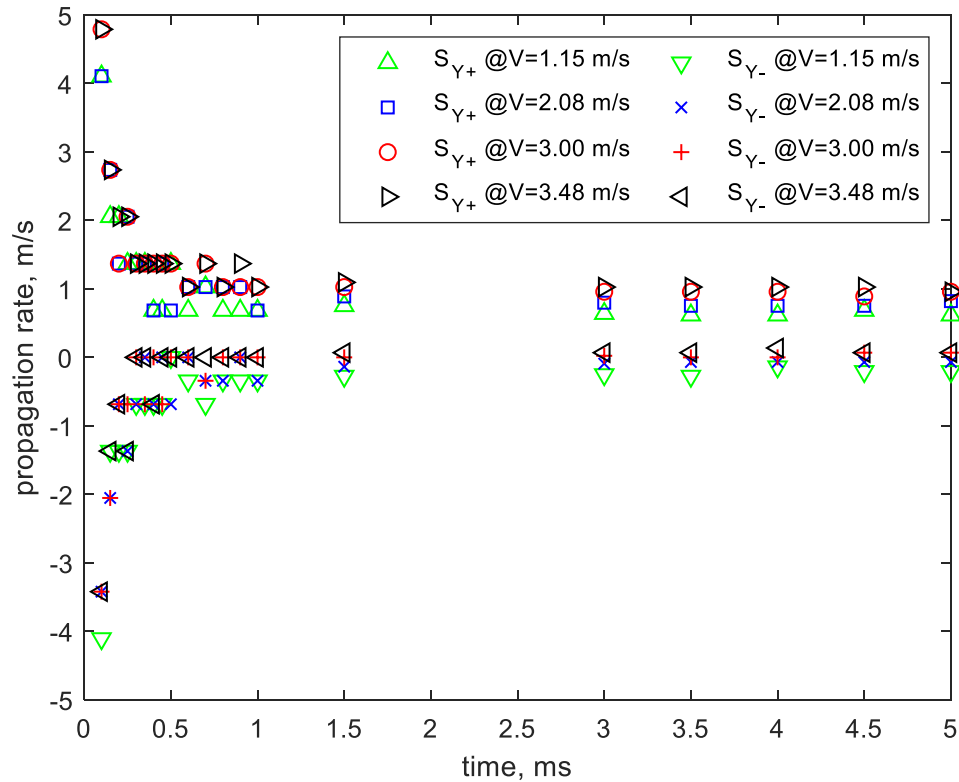
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Figure 5.22: Kernel width at varying velocity of flow

3

4 Figure 5.23 shows the effect of flow velocity on the flame front tip displacement
 5 along the flow axis. As shown, the general trend in propagation rates indicates
 6 decay from a highly stretched value to an asymptotic value irrespective of the
 7 velocity of the flow. The asymptotic values of S_{y+} were 0.63, 0.79, 0.95, 1.02 m/s at
 8 respective flow velocity U_0 values of 1.15, 2.08, 3.0, 3.48 m/s. Hence, an 80%
 9 increase in U_0 produce about 25% increase in S_{y+} . In contrast, the asymptotic
 10 values of S_{y-} were -0.25, -0.09, 0.02, 0.07 m/s at respective flow velocity U_0 values
 11 of 1.15, 2.08, 3.0, 3.48 m/s which represent a 64% decrease in S_{y-} for similar
 12 increase in flow velocities. The loss in propagation rates of the upstream flame
 13 front the result of higher aerodynamic strain on the flame kernel at higher flow
 14 velocities. The graphs also shows that a higher flow velocity had little or no
 15 influence on the propagation velocity of the flame front in the early stage up to
 16 about 0.5 ms. In addition, the the higher velocity flows were characterised by
 17 fluctuation in propagation rate before reaching the asymptotic values.



1

2

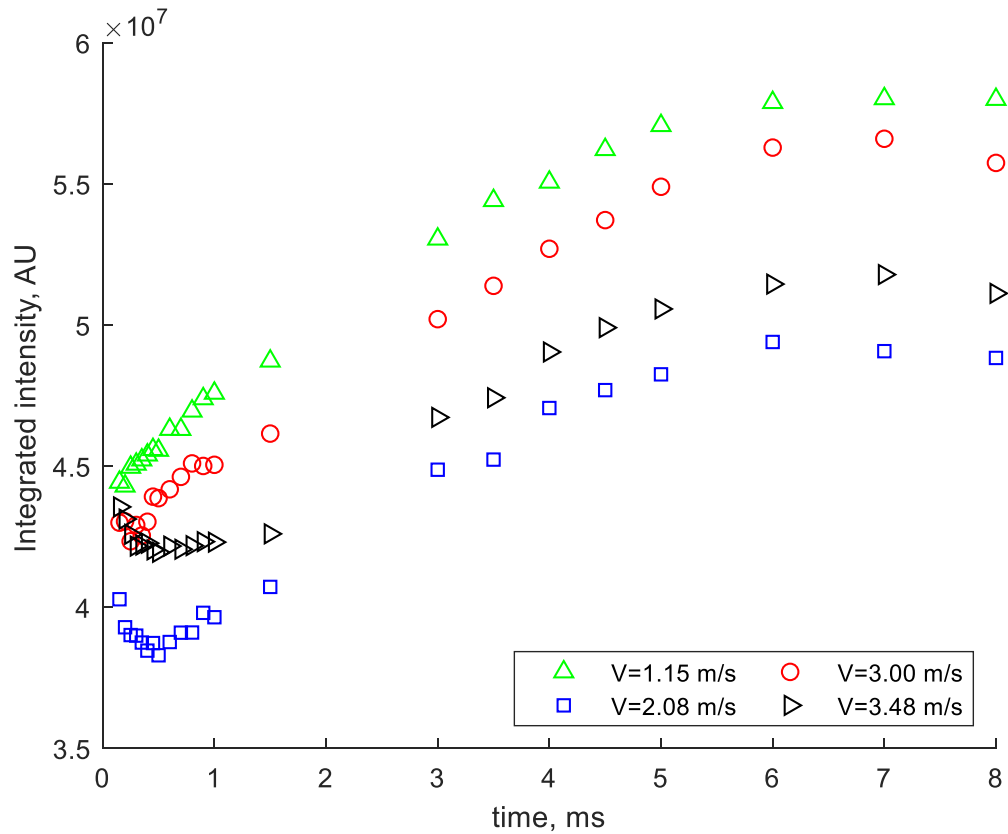
Figure 5.23: Flow velocity effect on S_{y+} and S_{y-}

3

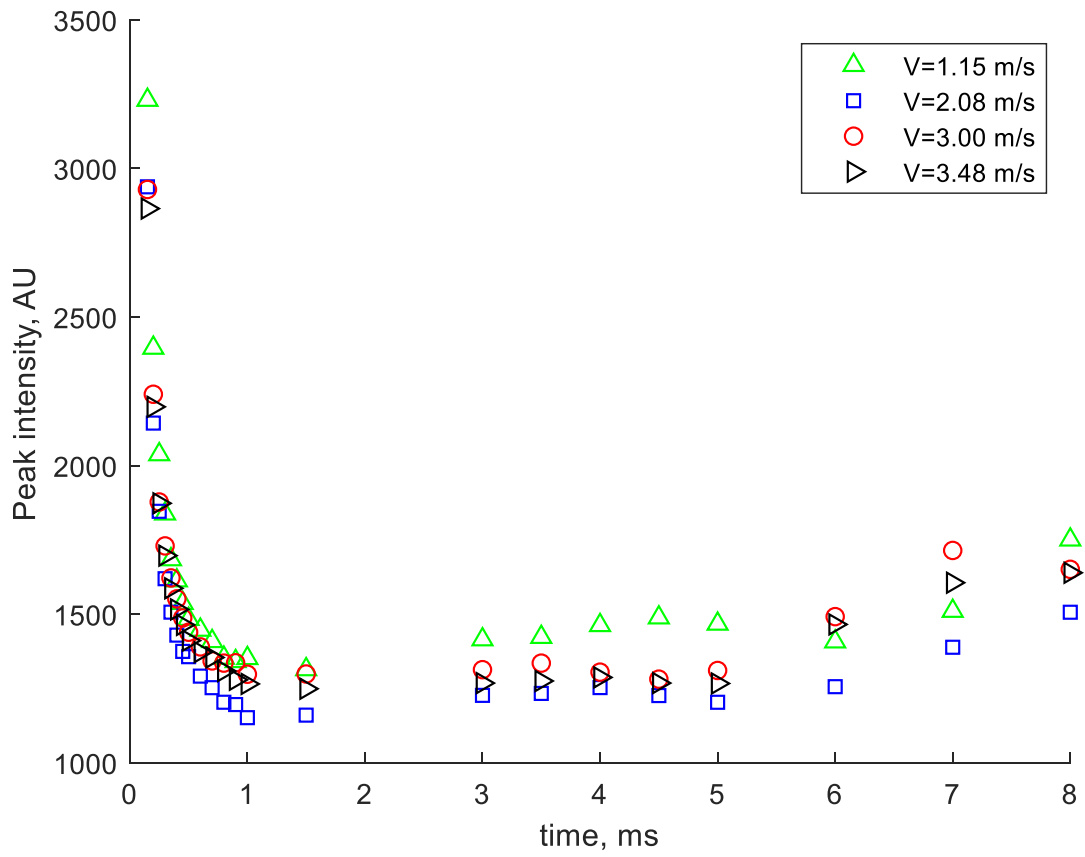
4 Figure 5.24 shows the evolution of spatially integrated OH* intensity for varying
 5 flow velocities. The observed trend for each flow velocity is similar to earlier
 6 observation with an initial decrease during chemical induction and a constant rise
 7 afterwards. From results of the integrated OH* intensities, it was difficult to
 8 determine the particular order of influence due to fluctuating output, hence a
 9 further investigation of the fluctuations from each ignition event is recommended.
 10 A unique observation from the graph is that the duration of chemical induction
 11 (which represents the time at which minimum value was obtained) tends to
 12 increase with an increase in flow velocity which might be interpreted as the result
 13 of cooling effect.

14 Similarly, the integrated OH* intensity, the observed trend in peak OH* intensity
 15 shown in Figure 5.25 did not provide adequate information on its sensitivity to
 16 increasing flow velocity. The inconsistency in the observed sensitivity of OH*

1 intensity could be linked to many factors. The first reason may be related to the
2 displacement effect of high flow velocities on location of the flame kernel centroid,
3 which means that a corresponding displacement in the observation window may
4 be necessary for a fair comparison. The second reason may be linked to possible
5 differences in spark efficiency due to the stochastic nature of breakdown process.



6
7 **Figure 5.24: Flow velocity effect on Integrated OH* intensity**
8



1

2

Figure 5.25: Flow velocity effect on Peak OH* intensity

1

2 5.6 Conclusions

3 The development of the flame kernel employing LISI has been characterized
4 experimentally in a lab-scale atmospheric burner with flowing stoichiometric
5 CH₄/Air mixtures. This was made possible through analysis of captured 2D
6 projection images resulting from the kernel OH* chemiluminescence. The major
7 findings are summarised below:

- 8 • Flame kernel visualisation using the OH* chemiluminescence revealed
9 similar geometric features observed in earlier from tomographic images. By
10 50 μ s, the kernel shape has evolved into a two-lobe symmetric toroid. This
11 was followed by the appearance of a front lobe before 100 μ s. Finally, a self-
12 sustained propagating flame ball resulted following the disappearance of
13 the front lobe.
- 14 • Analysis of the image frames following a geometric edge extraction showed
15 that the growth of the upstream flame front along the flow axis is
16 characterised by non-linear decay growth during the first 0.8 ms, followed
17 by a steady linear growth until it reached a maximum displacement of 12
18 mm in about 7 ms. On the other hand, the transverse growth of the flame
19 front towards the laser is characterised by relatively stable growth in the
20 first 1.5 ms, with sudden change in slope due to local quenching at the front
21 lobe and then a steady linear growth onwards. The resulting flame kernel
22 therefore differs in its spatial dimension (i.e., height greater than width)
23 which may be attributable to the asymmetry in the dimensions of the initial
24 spark kernel. Under the condition, it takes about 2 ms for the kernel height
25 (Y) to grow to a spatial scale about the size of the burner diameter (i.e., 22
26 mm) and kernel width reached a spatial scale about the size of the burner
27 diameter in just above 2 ms.

- 1 • Analysis of the flame front propagation rate along the three lobe directions
2 showed it decayed from a highly stretched value to a steady asymptotic
3 value close to the unstretched laminar flame speed. Based on these
4 observations, inference can be made that the initial kernel growth is the
5 result of both induced gas flows and chemical reactions while the steady
6 growth in the late kernels relies entirely on heat release due to chemical
7 reactions. It also highlighted the existence of three key development stages.
8 From a decaying kernel of hot gas, the kernel reached an initial steady
9 speed of about 11 m/s in 0.15 ms. This was the first phase of growth
10 dominated by induced gas velocity of the shock waves. A second decay
11 occurred between 0.2 and 0.25 ms, when the kernel reached a second stable
12 speed of 7.5 m/s due to formation of an igniting kernel. This marked the
13 second stage of development when the kernel propagates due to combined
14 influence of chemical reactions and induced gas velocity. The third and final
15 decay occurred between 0.8 ms beyond which a steady velocity of 3.5 m/s
16 was maintained. From this point onwards, the kernel propagated with
17 minimum velocity variation from its unstretched laminar value of 3.06 m/s.
- 18 • Further visualisation of the image colourmap revealed that during its
19 development, the reaction zone of the flame kernel (indicated by the OH*
20 intensity distribution) evolved from an isotropic to anisotropic flame front.
21 In the self-sustained flame, the reaction zones were concentrated at the
22 edges. These observations gave an idea of the different stages of the
23 development and the two-way interaction involving the flame-flow mixture
24 and flame-burnt products.
- 25 • Analysis of the luminous intensity of OH* intensity provided an indication of
26 the reactivity and heat release rate during development of the flame kernel.
27 Transition to a self-sustained flame kernel is characterized by decay in peak
28 OH* intensity to a minimum value which must be sustained for the kernel to
29 survive. The integrated OH* intensity initially decreased to a minimum
30 trough value which occurred at 0.2 ms in the base case study, followed by a

1 non-linear continuous rise to a maximum value in the fully developed flame.
2 The time elapsed before this minimum value was interpreted as the
3 chemical induction time which representing transition between a
4 quenching kernel of hot gas and formation of an igniting flame kernel. The
5 rising trend after an initial drop in integrated OH* intensity indicates that
6 the kernel is more likely to survive since cumulative heat release increased
7 during kernel expansion.

8 • A comparison of the kernel development at different incident energies (i.e.,
9 32, 64, 96 and 128 mJ) reveals that the early growth of the kernel is
10 enhanced by a higher energy, but this happens within a limiting threshold
11 energy. In the fully developed kernel however, the size of the kernel is not
12 influence by the energy. A comparison of the peak OH* intensities reveals
13 that the reactivity was unaffected by the energy of the incident beam except
14 in the first 0.5 ms. A comparison of the integrated OH* intensities shows
15 that the chemical induction duration increased for higher energies. An
16 interesting insight from this analysis is the fact that the maximum
17 integrated OH* intensity decreased greatly by 25% with energy increase
18 from 32 to 64 mJ, however, however, further increase in energy resulted a
19 reduction in the the negative effect. Based on the observations, the optimal
20 ignition energy should be decided based on the OH* production, however
21 further investigation with lower ignition energies is recommended. Further
22 details of the sensitivity analysis can be seen in Table 0.1 of Appendix E:
23 Sensitivity Analysis.

24 • A comparison of the kernel development at different bulk flow velocities
25 (i.e., 1.15, 2.08, 3.0 and 3.48 m/s) reveals and increased displacement of the
26 kernel centroid with higher flow velocity due to convective effects and as
27 well as an increased strain on the flame front upstream. Overall, there is no
28 remarkable increase in growth rate of the kernel due to increased velocity.
29 The combined effect of convection and increased turbulence at higher bulk
30 velocities resulted in a faster propagation of the flame front downstream.

- 1 There were fluctuations in both the peak OH* intensity and maximum
- 2 integrated intensity which made difficult to understand the influence of
- 3 increased velocity on reactivity. Also, the duration of chemical induction
- 4 appears to be longer for higher bulk velocity flows.

Chapter 6 Effects of fuel properties on the development characteristics of Laser-ignited Flame Kernel

6.1 Introduction

The demand for fuel flexibility and a shift towards lean combustion are the key motivations to study the fuel composition effect on laser-ignited flame kernels. The advantages of lean combustion are increased fuel efficiency and reduced NO_x emissions due to lower maximum combustion temperatures. Despite these benefits, combustion initiation under lean conditions is challenging and even after ignition initiation, incomplete combustion may result due to slow propagation. Hence, the need to understand the impact of leaning on combustion initiation and propagation. The desire for fuel flexible combustors is to increase energy sustainability. However, the impact of the wide variety of available fuel types and sources on the engine performance remain an issue of concern. Combustion variation is caused by differences in the thermochemical properties of fuels such as the laminar burning velocities (S_L), adiabatic flame temperatures (T_{ad}) as well as the Lewis Number (Le). It is therefore necessary to investigate the effects of these properties on the ignition dynamics.

In this chapter, the aim is to investigate the effect of the thermochemical properties of the fuel mixture on the development characteristics of the laser-ignited flame kernel. The study is divided into three main sections. In the first section, the composition of mixture is varied in terms of changing equivalence ratios of methane/air mixture which also represents changing values of S_L or T_{ad} . In this investigation, the methane/air mixtures were selected to include flows within the laminar and turbulent flow regimes. In the second section the mixture composition is varied in terms of changing fuel types with constant adiabatic flame temperature while in the third section different fuel types with constant S_L are compared. In both sections two and three, differences in flame kernel

characteristics are viewed from the angle of differences in the fuel type since either a constant S_L or T_{ad} is maintained. These studies were achieved using the same experimental setup as in chapter 5. The major difference is the introduction two new fuels (i.e., propane and ethylene) in addition to methane.

6.2 Effects of change in fuel mixture composition

In this section, the effect of changing fuel-air ratio on the development characteristics of methane flame kernel is reported. Three ϕ values (1.0, 0.9 and 0.8) were selected in the laminar flow region and another three ϕ values (1.0, 0.9 and 0.8) in the turbulent flow region making a total of six test cases. The corresponding T_{ad} values are 2226 K, 2134 K and 1996 K respectively[110]. Table 6.1 is summary of the selected flow conditions and the properties. As shown in the table, the compared mixtures all have nearly the same bulk flow velocity. In addition, all experimental conditions except mixture ratios were kept constant.

Table 6.1: Summary of flow conditions with varying equivalent ratios

Test Case	ϕ	Flow Rate (lpm)		U_0 (m/s)	Re (Pa.s)	T_{ad} [110] (K)	u_L [112] (cm/s)	$\frac{\rho_u}{\rho_b}$
		Air	CH ₄					
Laminar flows	1	23.8	2.5	1.15	1694	2226	41	7.47
	0.9	24.3	2.3	1.17	1718	2134	36	7.16
	0.8	25.0	2.1	1.19	1750	1996	29	6.70
Turbulent flows	1	101.0	10.6	4.89	7184	2226	41	7.47
	0.9	101.6	9.6	4.87	7169	2134	36	7.16
	0.8	102.4	8.6	4.87	7165	1996	29	6.70

Figure 6.1 visualizes the effect of reducing fuel/air ratios on flame kernel evolution from the mean OH* chemiluminescence images. By comparing mean images acquired at the same ignition delays, the developing kernels showed no significant

difference in their sizes except that the leanest mixture was more stretched leading an earlier quenching at the front lobe. At the developed stage, the kernel sizes were clearly distinct from each other as the kernel become smaller at lower ϕ values. A similar trend is observed with the kernels developing in turbulent mixture environment as shown in Figure 6.2. As shown by the image colour scales, the OH* intensity at the kernel boundaries are more clearly separated from that of the hot gases in the stoichiometric mixture than the lean mixtures while in the turbulent FF, the OH* intensity seem to indicate a wider thickness as the mixture equivalence ratio decreases.

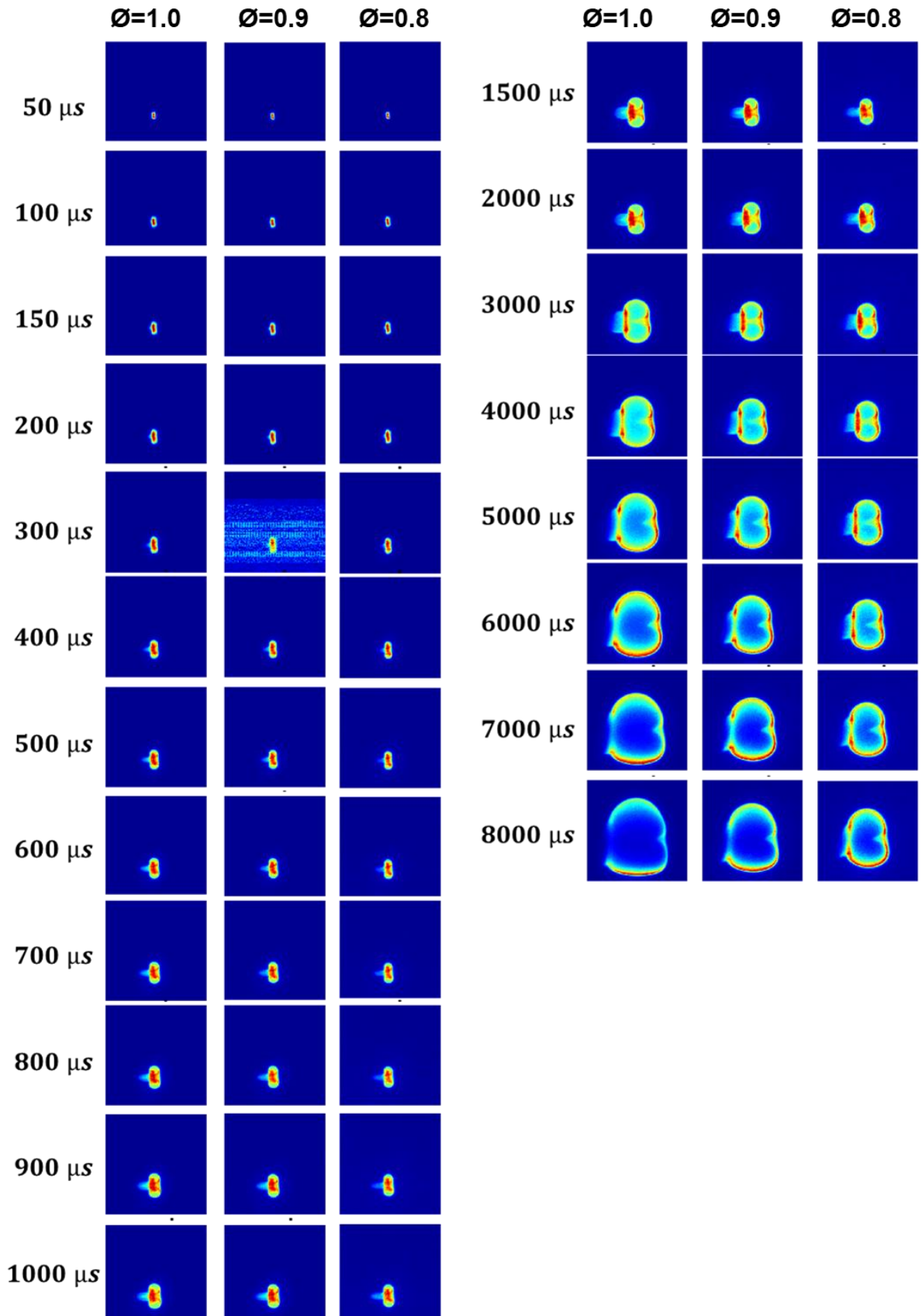


Figure 6.1: Equivalence Ratio Dependence of Flame Kernel in Laminar Mixtures (FOV: 56X56 mm²)

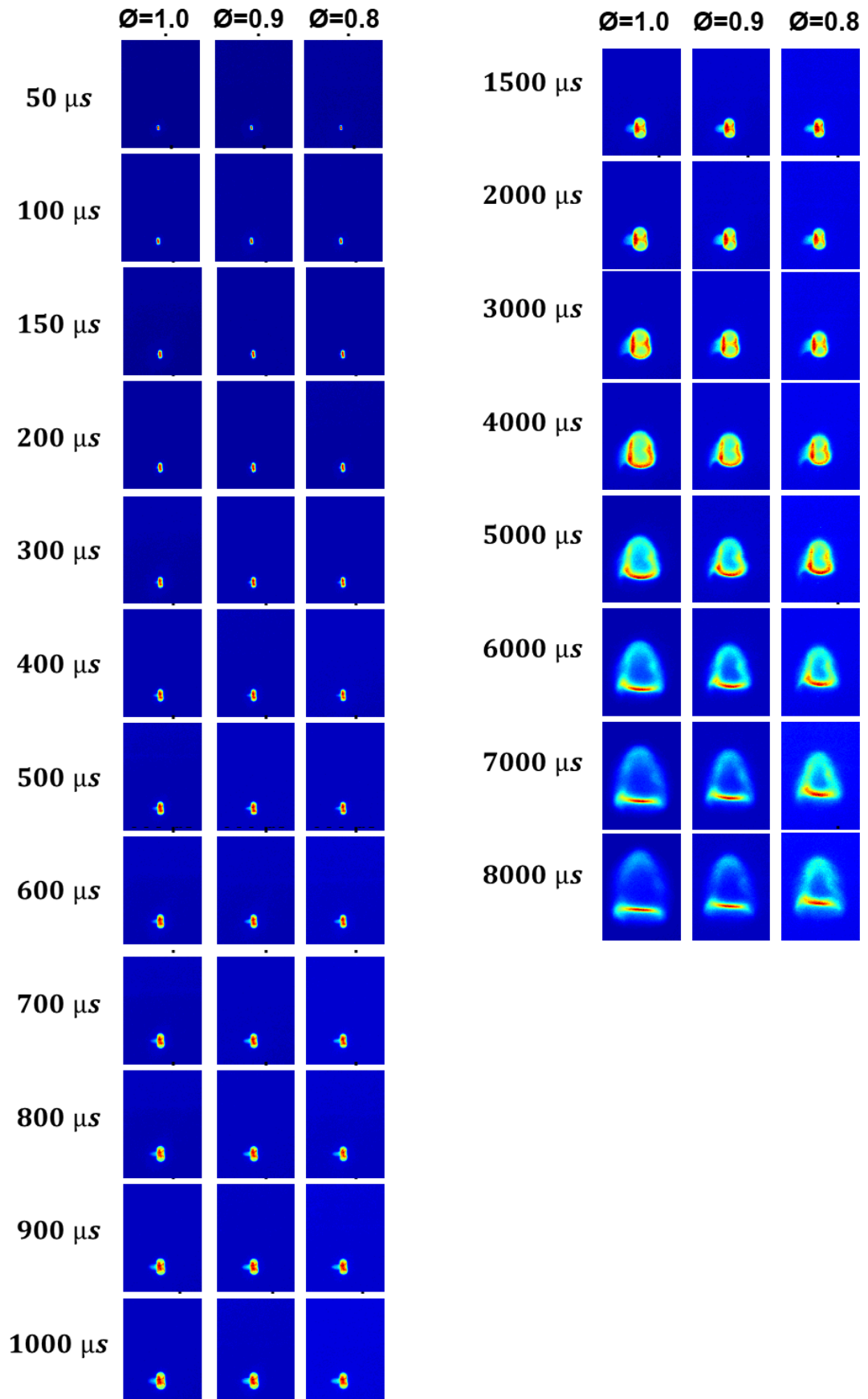


Figure 6.2: Equivalence Ratio Dependence of Flame Kernel in Turbulent Mixtures (FOV: 56X56 mm²)

Figure 6.3 - Figure 6.4 compares the observed peak OH* intensities with changing equivalence ratios ($\phi=1.0, 0.9, 0.8$) for different CH₄/Air mixtures. As in previous observations (see Chapter 5), the peak OH* intensity for each mixture follows an asymptotic trajectory which indicates cooling rate resulting from an imbalance between production and consumption of the OH* radicals. In the present investigation, the general trend is that higher OH* intensity peak is synonymous with higher adiabatic temperature which is dependent on the mixture equivalence ratio. In the self-sustained flame kernels (1.5 – 10 ms) obtained for laminar mixtures, the highest average peak of 1400 AU is obtained in the stoichiometric mixture (i.e., $\phi=1.0$) while lowest average peak of 800 AU is obtained in the stoichiometric mixture (i.e., $\phi=0.8$). This indicates higher reactivity for near stoichiometric mixtures (i.e., $\phi=1.0$ and $\phi=0.9$) due to higher adiabatic flame temperatures and lower reactivity in lean mixtures (i.e., $\phi=0.8$) due to lower adiabatic flame temperatures. By comparing peak OH* intensities of the three turbulent flow mixtures, the same trend is observed as in the laminar cases having the same equivalent ratios but slightly lower reactivity. This relationship between equivalent ratio and peak OH* intensity is consistent with previous studies [50],[158],[159] where reactivity is associated with the rate of heat release. An interesting result from the sensitivity analysis is that the variation is linear in laminar flows but non-linear in the turbulent case.

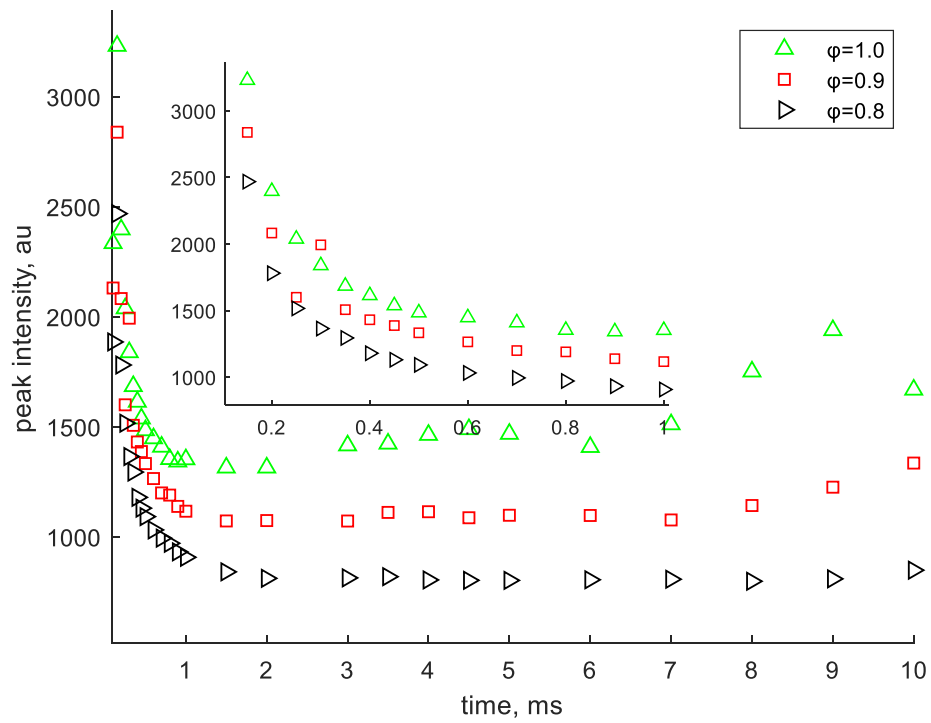


Figure 6.3: Peak OH* Intensities at varying Equivalence Ratios (Laminar Mixtures)

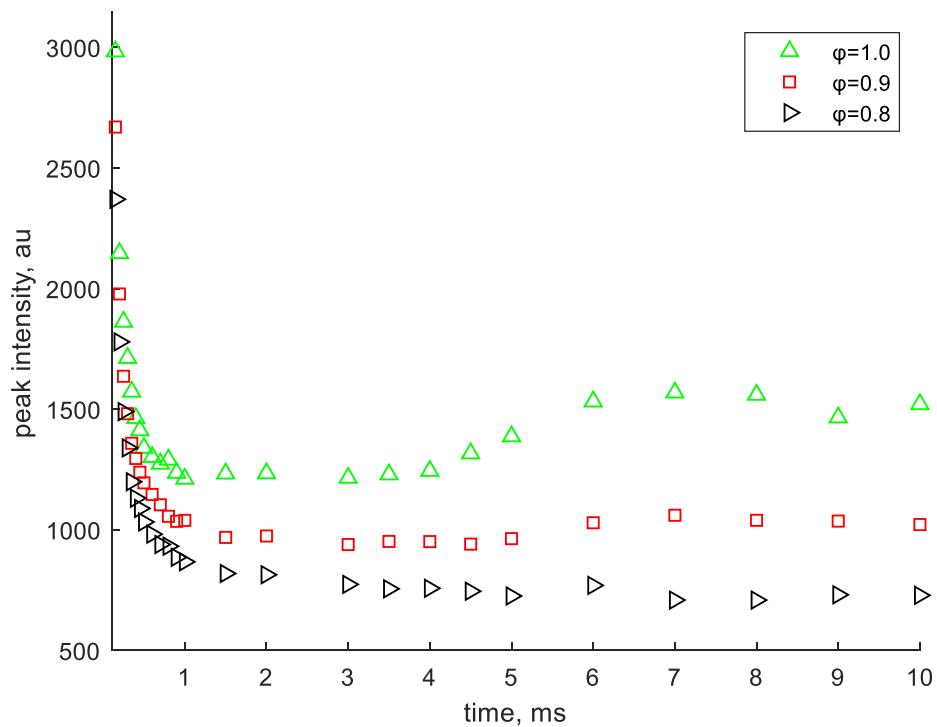


Figure 6.4: Peak OH* Intensities at varying Equivalence Ratios (Turbulent Mixtures)

Figure 6.5 - Figure 6.6 compares the spatially integrated OH* intensity with equivalence ratios ($\phi=1.0, 0.9, 0.8$) for different CH₄/Air mixtures. For the laminar mixtures, the initial order of magnitude of integrated OH* intensity did not give any indication, however the time of transition from a decreasing trend to an increasing trend happened in the following order: $\phi=1.0 < \phi=0.9 < \phi=0.8$. This may be an indication that chemical induction duration is shorter at $\phi=1.0$ (i.e., 0.2 ms) and longer at $\phi=0.8$ (i.e., 0.45 ms). Following ignition, the relative increase in production level of OH* radical was highest for the mixture with $\phi=1.0$ and least for the mixture with $\phi=0.8$ as indicated by the slope of the graphs. Similar trend is observed in the turbulent flows shown in Figure 6.6 with chemical induction being completed at 0.35 ms for the near stoichiometric mixtures while that of $\phi=0.8$ was completed after 0.4 ms. From the plots, the initial order of integrated OH* intensity was $1.0 < \phi < 0.8$, however the relative production rate increased in the reverse order (i.e., $1.0 > \phi > 0.8$) until they converged at 2 ms. Beyond 2 ms, the effect of increase in fuel-air ratio was clearly marked by higher OH* intensity as well as an increasing rate of OH* production until about 6 ms when no further increase in relative OH* intensity could be observed. The sensitivity analysis further shows the variation is linear in laminar flows but non-linear in the turbulent case.

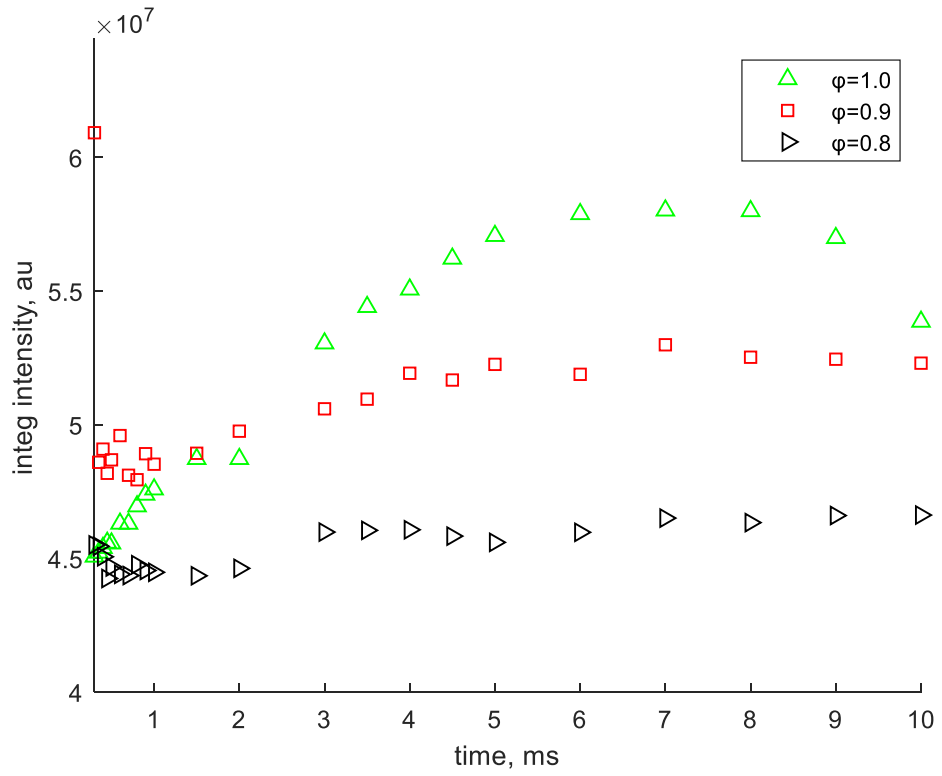


Figure 6.5: Integrated OH* Intensities at varying Equivalence Ratios (Laminar Mixtures)

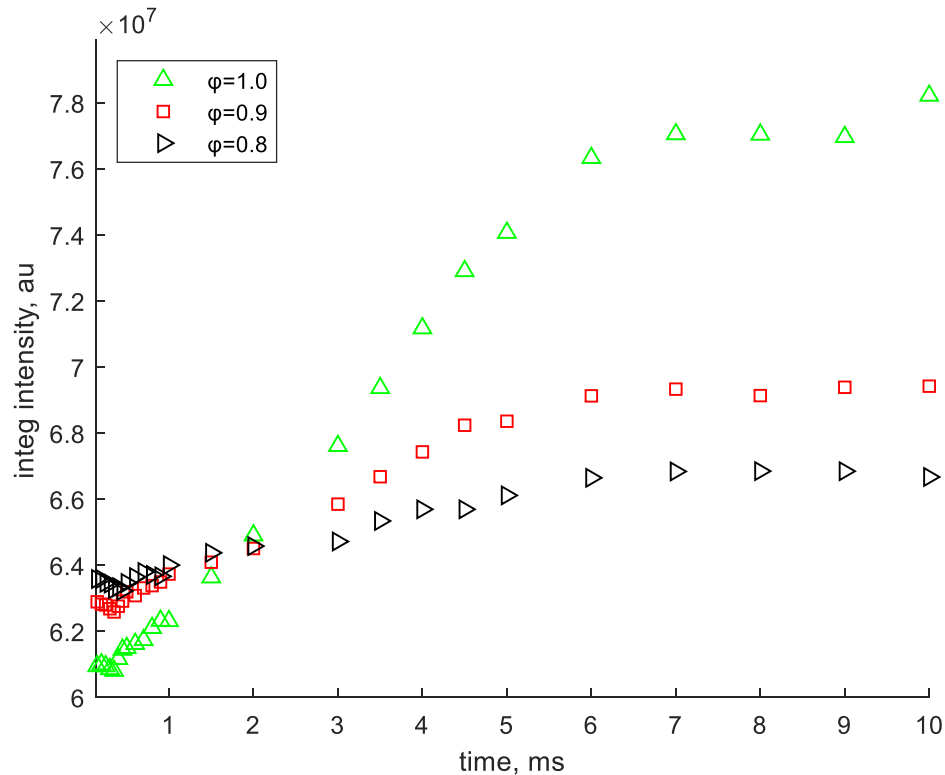


Figure 6.6: Integrated OH* Intensities at varying Equivalence Ratios (Turbulent Mixtures)

Figure 6.7 - Figure 6.8 compares the evolution of the axial flame tip locations with ϕ values (i.e., 1.0, 0.9, 0.8) for different CH₄/Air mixtures. A common trend from both plots is that the FF tip displacement (in both upstream and downstream directions) decreased from $\phi=1.0$ to $\phi=0.8$ and this was more prominent in the late developed stage (i.e., above 1 ms). According to Figure 6.7, the leanest flame kernel required 9 ms to travel 28 mm upstream compared to 7 ms required by the stoichiometric flame. An obvious distinction between both plots is that in the laminar flows the growth is nearly symmetric about the laser axis while in turbulent flows the growth symmetry is lost. The asymmetry in kernel growth under turbulent environment is due to convective effect of high flow velocity which results in a displacement of the kernel centroid from the initial location. It is also seen that the impact of high flow velocity is more prominent in lean mixtures than the stoichiometric. Further investigation of the 2D-scatter plot (see Figure 6.9) of the laminar mixtures did not show any peculiar variation in the horizontal location of the axial FF tip. The plot shows that stabilization of the upstream flames

produced in all the mixtures would more likely stabilize in the direction away from the laser source while the downstream flame would stabilize if the flame holder were placed along a horizontal axis as the ignition point. Unlike the laminar flows, the turbulent flow cases (Figure 6.10) show a tendency to blow-out, however the trends remain similar.

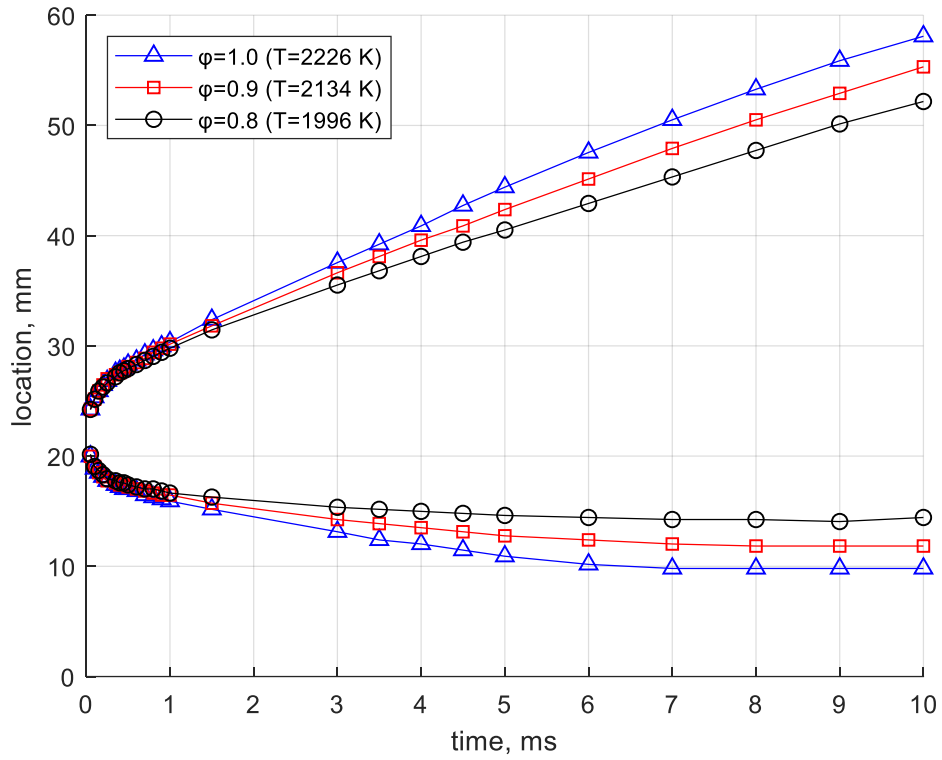


Figure 6.7: Axial Flame Front Locations at varying Equivalence Ratios (Laminar Mixtures)

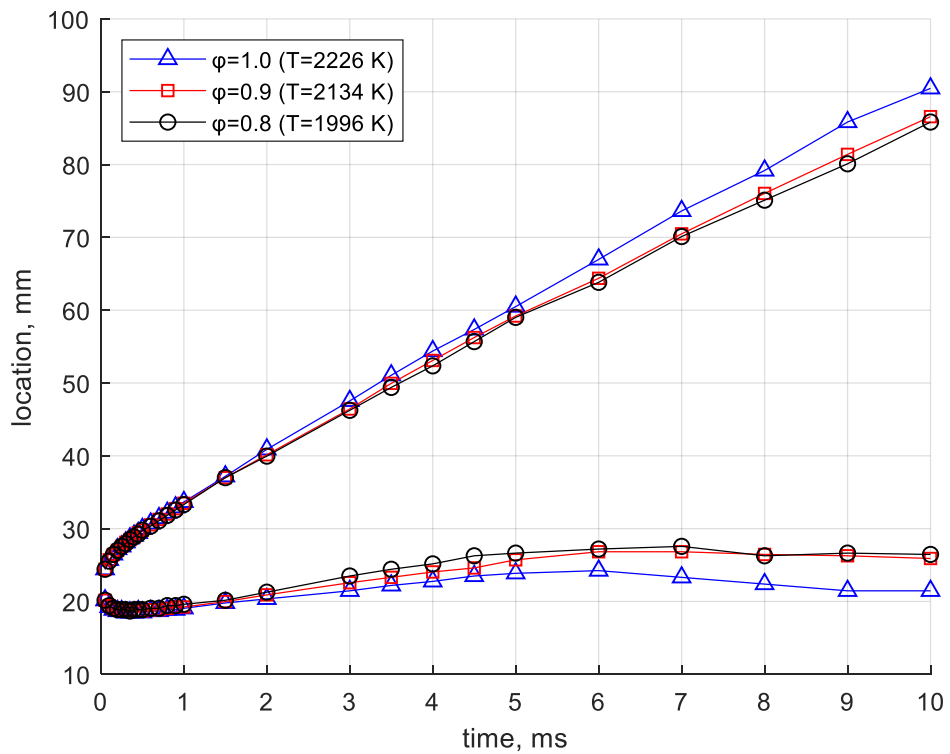


Figure 6.8: Axial Flame Front Locations at varying Equivalence Ratios (Turbulent Mixtures)

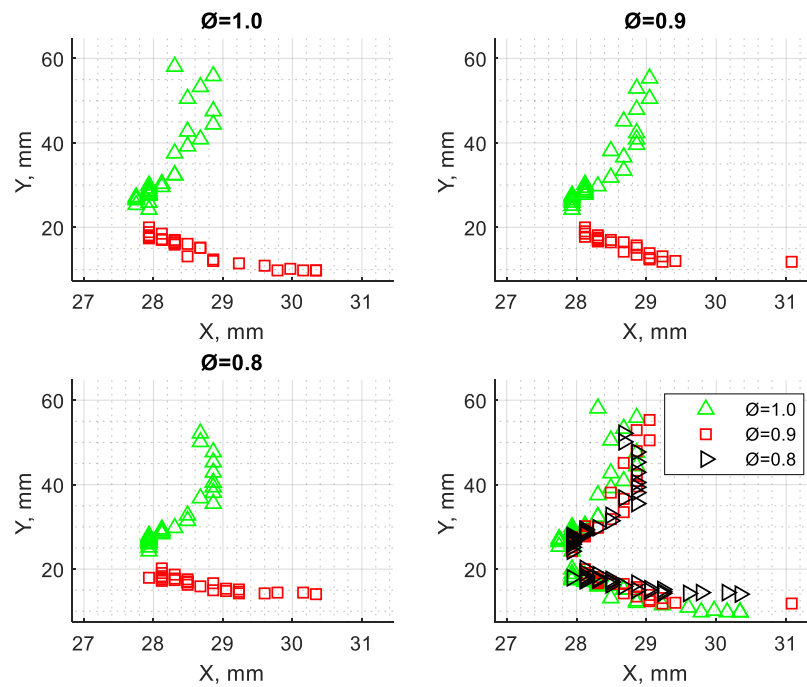


Figure 6.9: 2D Scatter Plot of Axial Flame Front Locations at varying Equivalence Ratios (Laminar Mixtures)

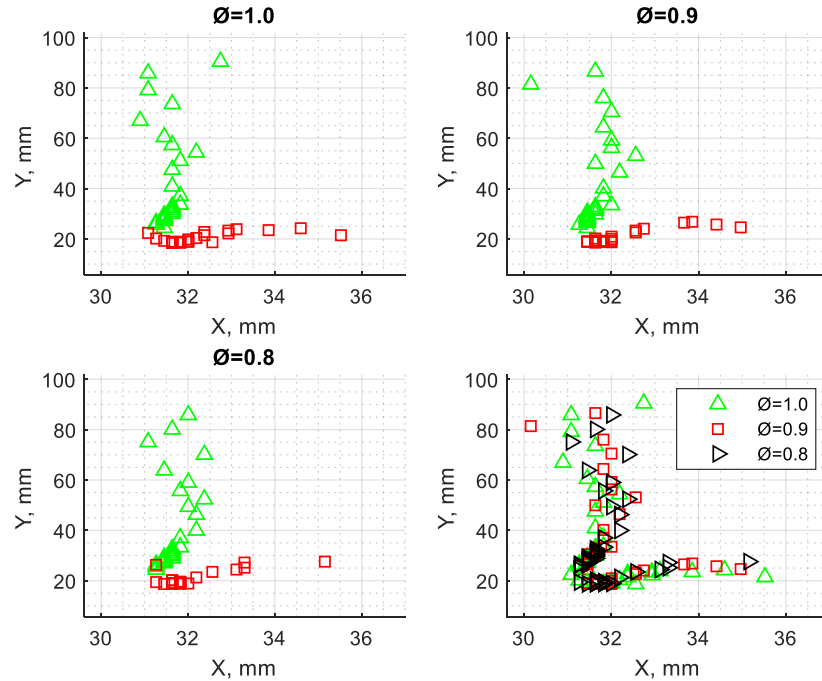


Figure 6.10: 2D Scatter Plot of Axial Flame Front Locations at varying Equivalence Ratios (Turbulent Mixtures)

Figure 6.11 - Figure 6.12 shows the variation of flame front propagation speed (S_y) with equivalent ratios (i.e., $\phi = 1.0, 0.9, 0.8$) in CH_4/Air . The general trend is that S_y decreases asymptotically from a highly stretched initial propagation speed to a value close to the laminar growth rate (S_L). S_L is calculated from the product of the unstretched laminar flame velocity (u_L) and the expansion coefficient ($\frac{\rho_u}{\rho_b}$):

$$S_L = \frac{\rho_u}{\rho_b} * u_L \quad 6.1$$

where: ρ_u and ρ_b are the densities of the unburnt and burnt gas, respectively.

It is also evident from Figure 6.11 that the flame kernel development to a stable flame front evolved in three key transition stages as observed previously. For example, in the laminar mixture with $\phi=0.9$, the first transition (which results from the front lobe development) was marked by stable S_{y+} value of 11 m/s at 200 μs , while the second transition (which marks the end of chemical induction) was indicated by another stable speed of 4 m/s at 350 μs . In the last stage of development, which began at 1.5 ms, a steady state value of 2.96 m/s was

maintained. Contrary to expectations from mixtures of different S_L values, there was no noticeable variation S_{y+} with changing ϕ until about 1.5 ms. The same trend is observed in the kernel development for the turbulent mixtures as shown in Figure 6.12. This behaviour could be linked to the different level of stretch experienced by different mixture flame kernels. From 1.5 ms onwards, the variation of propagation speed with equivalence ratio occurred in the correct order of magnitude as the laminar unstretched growth rate, S_L of each mixture. A similar trend is observed in the turbulent flow mixtures as shown in in Figure 6.12. Just like the laminar flow mixtures, the decay from a high initial velocity to a more stable velocity involved two transition phases before reaching a stable velocity of about 7 m/s within 350 μ s. Unlike the laminar mixtures, all the three equivalence ratios were marked by velocity fluctuations between 7 m/s and 11 m/s from this point until 700 μ s before travelling steadily at 7 m/s. There were also no noticeable differences in the magnitude of S_y until 1.5 ms when it decreased due to reduction in ϕ values. In addition, S_{y+} was marked by some degree of fluctuations unlike the laminar mixtures.

To better understand the relative variation in growth rate with equivalence ratios, the axial flame front propagation speed is normalised by the unstretched laminar flame speed. Figure 6.13 - Figure 6.14 compares the normalised propagation speed of the axial flame front with the corresponding equivalent ratios (i.e., $\phi = 1.0, 0.9, 0.8$) in laminar and turbulent mixtures, respectively. The deviation in S_y from the dashed lines gives the measure of the stretch experienced by the flame front. The Geometric Stretch (K) values for an outwardly propagating spherical flame of radius (r), can be evaluated using the below equation [90]:

$$K = \frac{2}{r} * \frac{dr}{dt} \quad 6.2$$

In the present case, r could be replaced by Y_+ or Y_- while $\frac{dr}{dt}$ could be replaced with S_{Y+} or S_{Y-} . Analysis of the downstream flame front shows that the propagation rate, S_{y+} decreased from an initially stretched value until it converges to a steady value close to the unstretched laminar value, S_L . For the kernels propagating in laminar mixtures, measurement reveals that the most stretch occurred in the leanest mixture with adiabatic flame temperature of 1996 K (i.e., S_{Y+} @ $0.8\phi =$

$1.33S_L$) while the least stretch occurred in the stoichiometric mixture with adiabatic flame temperature of 2226 K (i.e., S_{y+} @ $1.0\phi = 1.08S_L$). A similar trend is observed in the flame kernels propagating in turbulent mixtures but with negative stretch impact such that the normalised S_{y+} values at steady state lie below the 1.0 line. Analysis of the upstream flame front shows that the propagation rate, S_{y-} continues to decrease after reaching the unstretched value S_L before converging to its asymptotic values. In both laminar and turbulent flame kernels, the stretch sensitivity to leaning appear in the reverse order with the leanest mixture being the most negatively stretched.

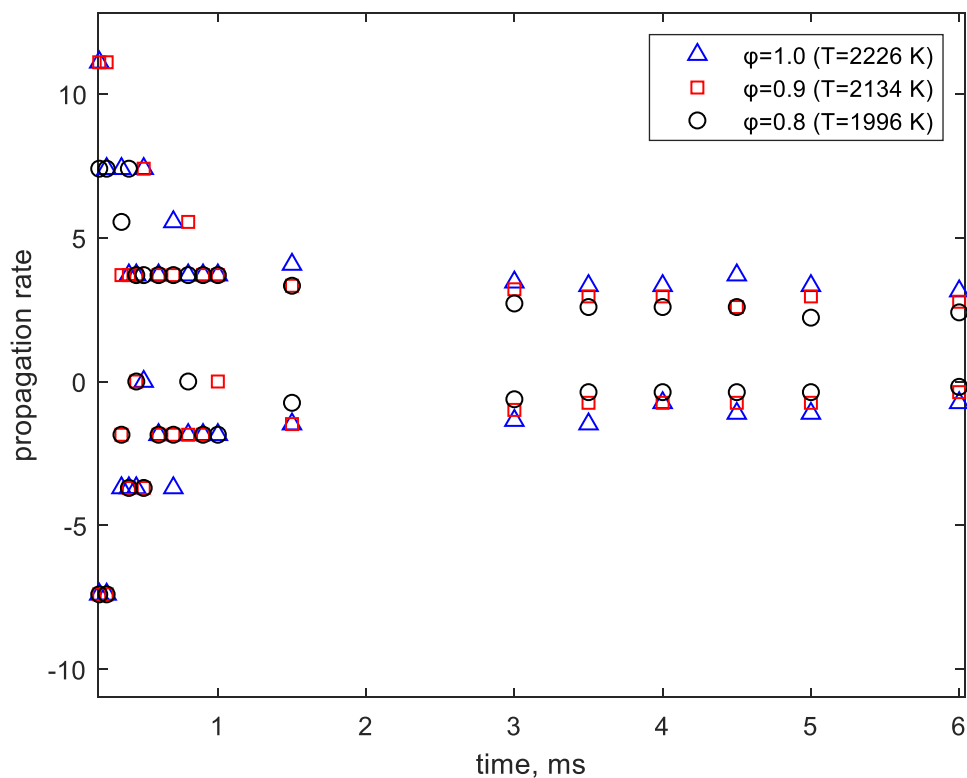


Figure 6.11: S_{y+} for varying ϕ (Laminar Mixtures)

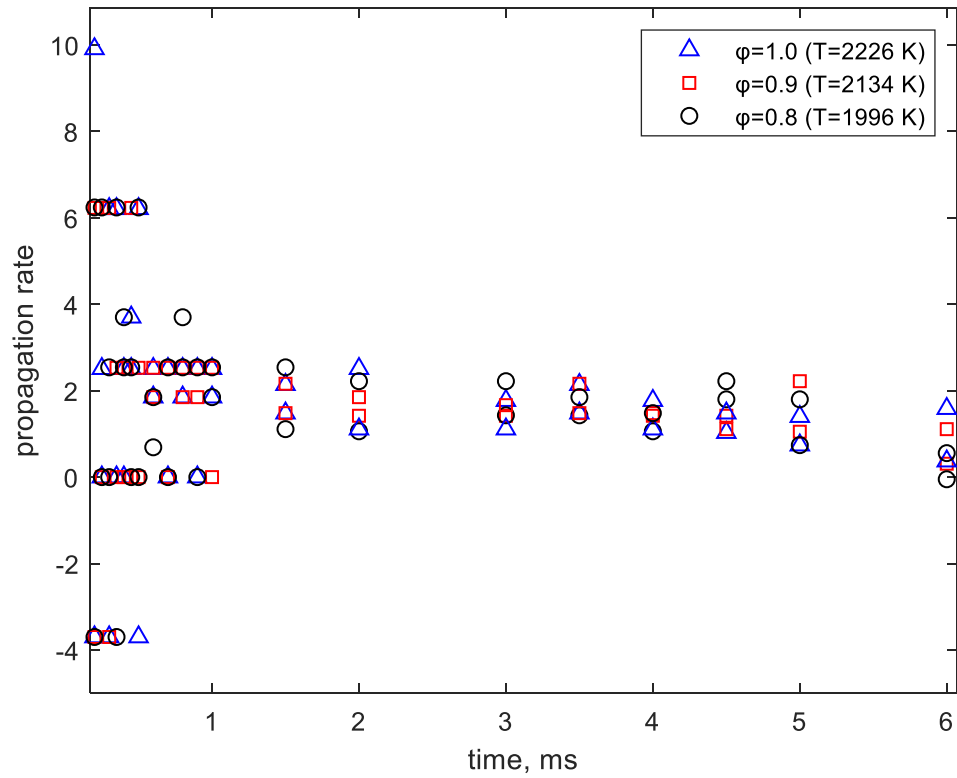


Figure 6.12: S_{y+} for varying ϕ (Turbulent Mixtures)

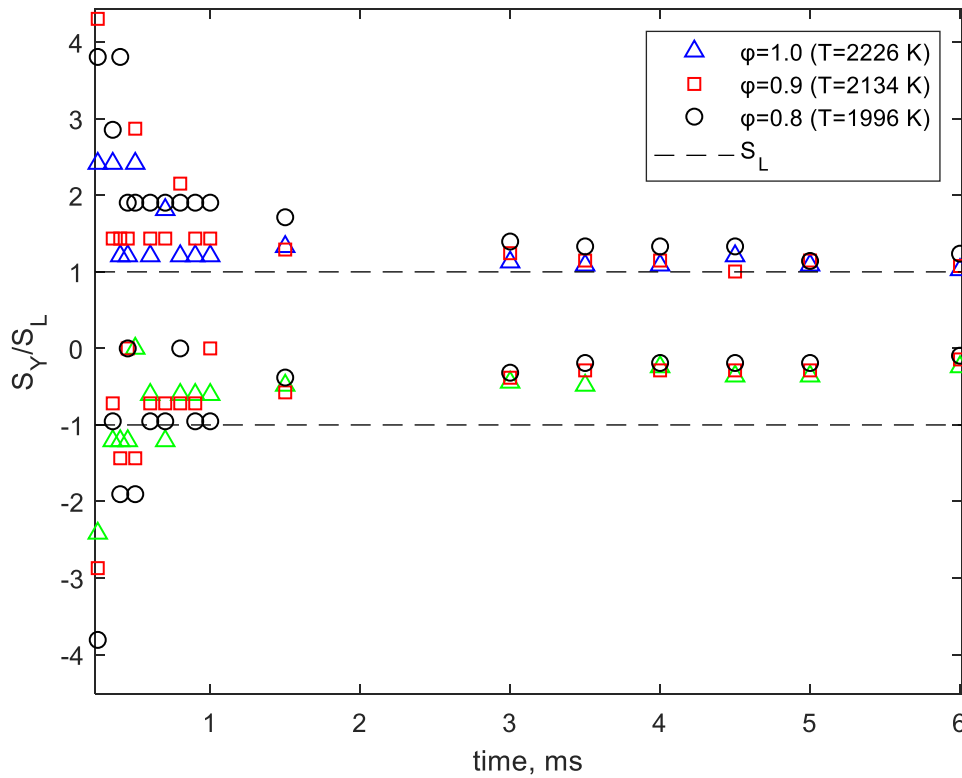


Figure 6.13: S_{y+}/S_L for varying ϕ (Laminar Mixtures)

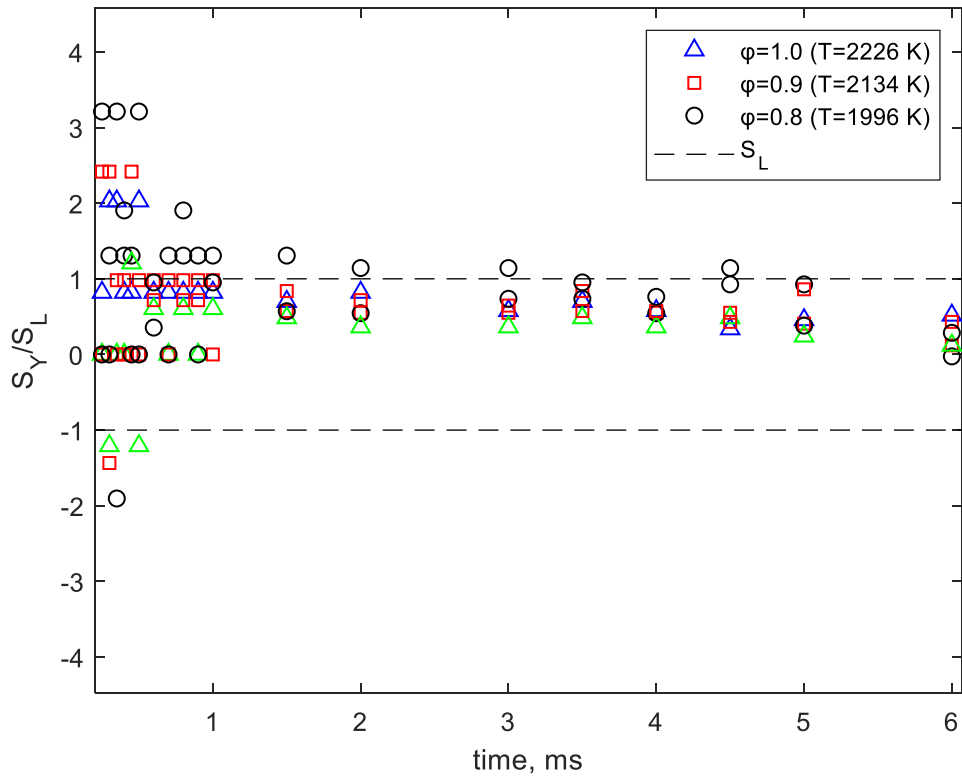


Figure 6.14: S_{y+}/S_L for varying ϕ (Turbulent Mixtures)

6.3 Fuel effects based on constant adiabatic flame temperature.

It is well known that the adiabatic flame temperature, T_{ad} through the Arrhenius kinetics exerts a dominant influence on the burning velocity since it is directly linked with the heat of combustion of the fuels [109]. Therefore, a means to experimentally determine the actual influence of the selected fuel on the propagation of the flame is by comparing fuel/air mixtures with different chemical composition but constant flame temperature. In this section, the effect of fuel on the flame kernel characteristics of three gaseous fuels (methane, propane and ethylene) with constant T_{ad} is investigated. The investigation is approached systematically from two angles. First, three laminar mixtures (i.e., CH_4/air @ $\phi=1.0$, C_3H_8/air @ $\phi=0.94$ and C_2H_4/air @ $\phi=0.84$) with constant T_{ad} (i.e., 2226 K) were first selected and their kernel characteristics compared to quantify the differences due to fuel type. This is followed by further investigation of the observed flame kernel characteristics under leaner mixtures (i.e., CH_4/air @ $\phi=0.9$, C_3H_8/air @ $\phi=0.86$ and C_2H_4/air @ $\phi=0.78$) with a constant reduced T_{ad} (2134 K). Overall, a total of six (6) flow conditions were compared as given in Table 6.2. The mixture ratios were selected based on the T_{ad} vs. ϕ plot obtained using GASEQ chemical equilibrium software [110] as shown in Figure 6.15. Besides T_{ad} being constant, the mixtures were selected such that all experimental conditions including the bulk flow velocity were kept constant.

Table 6.2: Summary of selected Laminar Flow conditions based on constant T_{ad} .

Test Case	ϕ	Flow Rate (lpm)				U_0 (m/s)	Re (Pa.s)	T_{ad} [110] (K)	u_L [112] (cm/s)	$\frac{\rho_u}{\rho_b}$
		Air	CH ₄	C ₃ H ₈	C ₂ H ₄					
Laminar mixture @ 2226 K	1.00	23.8	2.5	0.00	0.00	1.15	1694	2226	41.0	7.47
	0.94	25.3	0.0	1.00	0.00	1.17	1718		41.6	8.71
	0.84	25.5	0.0	0.00	0.90	1.19	1750		51.8	7.47
Laminar mixture	0.9	24.3	2.3	0.00	0.00	1.17	1718	2134	36.0	7.16
	0.86	26.3	0.0	0.95	0.00	1.19	1859		37.2	8.35

@ 2134										
K	0.78	25.9	0.0	0.00	0.85	1.17	1777		45.0	7.16

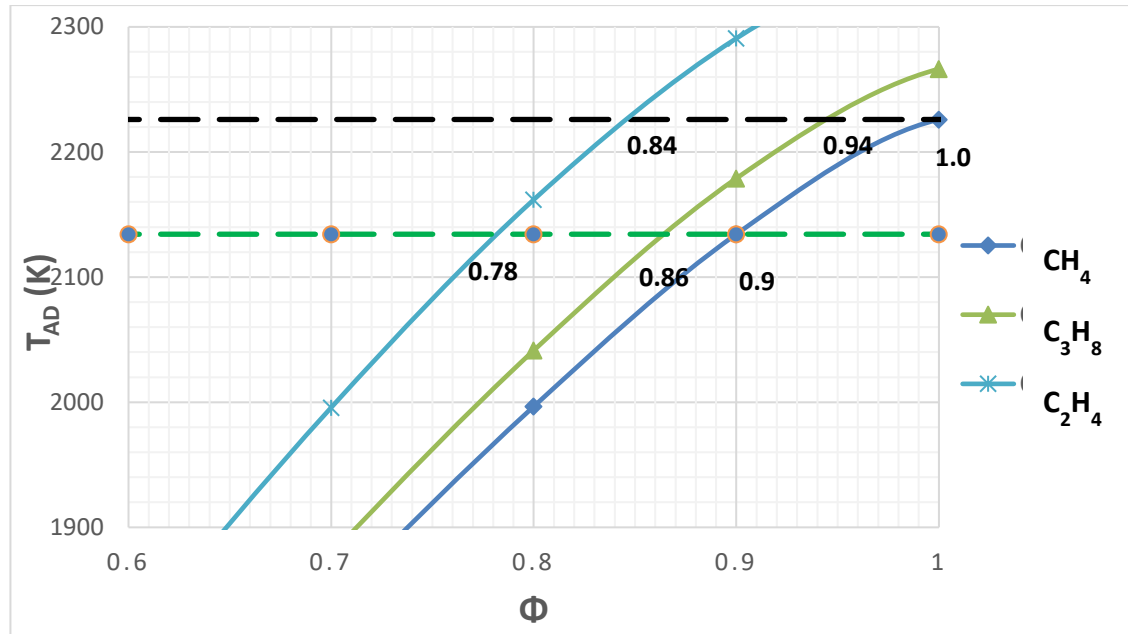


Figure 6.15: Selected flow conditions on $T_{ad} - \phi$ plots [110].

Figure 6.16 are sequences of mean OH^* chemiluminescence images which visualizes the comparative characteristics of the three laminar fuel/air mixtures (i.e., CH_4/air @ $\phi=1.0$, $\text{C}_3\text{H}_8/\text{air}$ @ $\phi=0.94$ and $\text{C}_2\text{H}_4/\text{air}$ @ $\phi=0.84$) with the same T_{ad} value of 2226 K. As expected, all the flame kernels exhibit common geometric features in their development (i.e., evolved from a two-lobe symmetric toroid to a three-lobe asymmetry and finally to a flame ball). However, there are significant differences in their growth which is obvious from 1 ms onwards. The first observation from the sizes of the late kernels is that propane experiencing the slowest growth while methane experienced the fastest growth. Another difference shown by the images is that between 0.5 and 3 ms, the front lobe of the propane flame kernel was the most stretched leading to an early local quenching which may have affected its growth rate compared to those of methane and ethylene. Comparing the flame kernels of both methane and ethylene, it appears that the difference in growth rate is mostly significant in the upstream flame front with higher strain effect on ethylene flame than the methane flame.

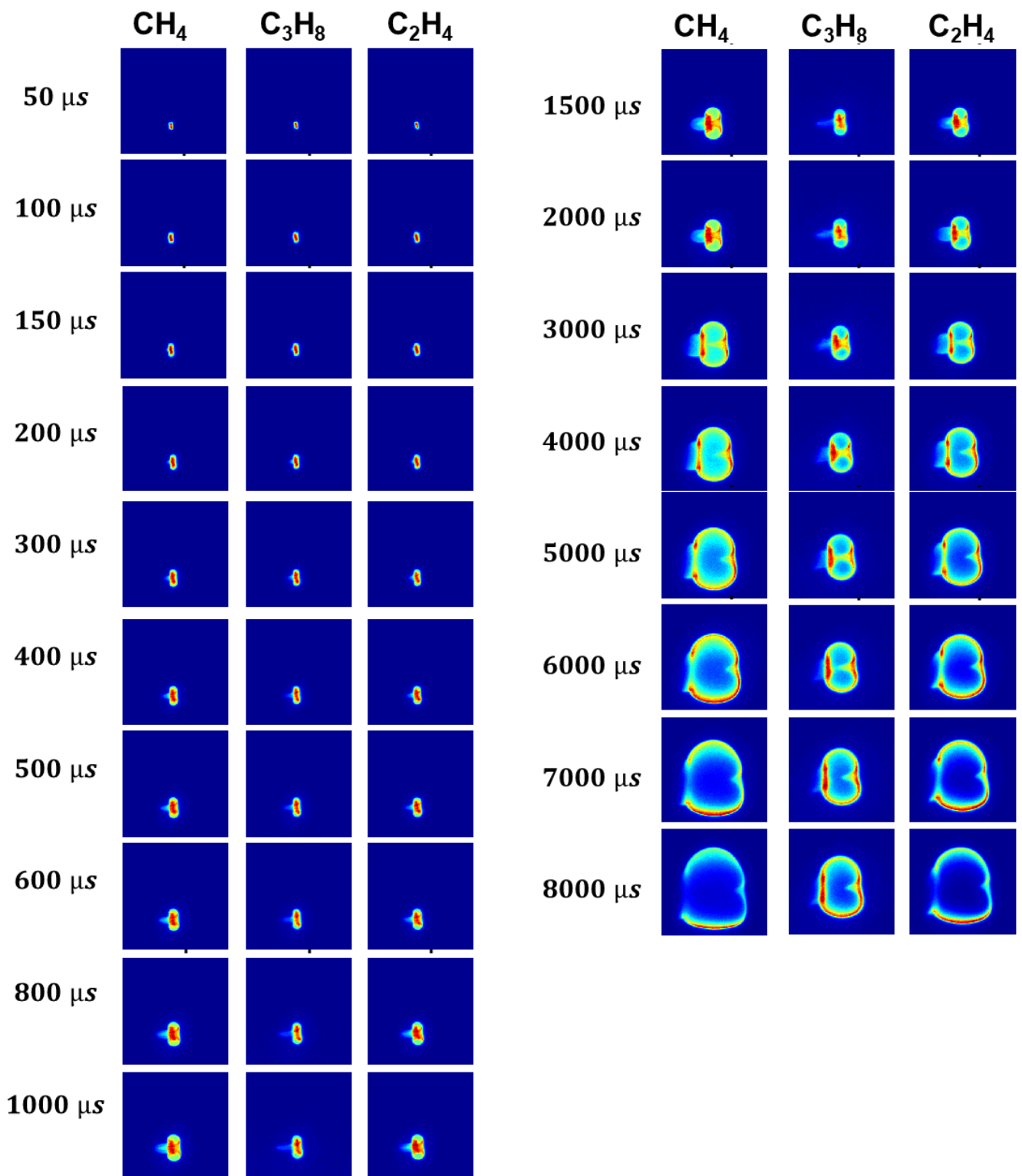


Figure 6.16: Visualization of Flame Kernel Development in mixtures at 2226 K (Image FOV: 56x56 mm²)

Figure 6.17 - Figure 6.18 compares the peak OH* intensities obtained from the flame kernels of the three fuels at 2226 K and 2134 K respectively. Similar to the results reported in section 6.2, the peak intensity of the flame kernel for each fuel dropped at lower adiabatic flame temperature. The main observation from both graphs is that for the same adiabatic temperature, the order of peak intensity values is $C_2H_4 > CH_4 > C_3H_8$. Therefore, it can be inferred that ethylene produced the most reactive flame kernel while propane produced the least reactive flame kernel. Although the CH_4 and C_3H_8 kernels has similar initial values at 150 μs , the CH_4 -flame kernel recovered more quickly from the initial rapid cooling, thus its peak intensity became higher than the C_3H_8 flame kernel. Based on the C/H ratios, it is expected that methane with lower C/H will be more reactive than propane just as observed. While the C/H may be higher for ethylene than the rest, the unstable double bonds present no doubt played a role in its high reactivity. In addition, the slow reactivity of the propane kernel could be associated with heat losses due to excessive stretching of the front lobes which persisted until 2 ms.

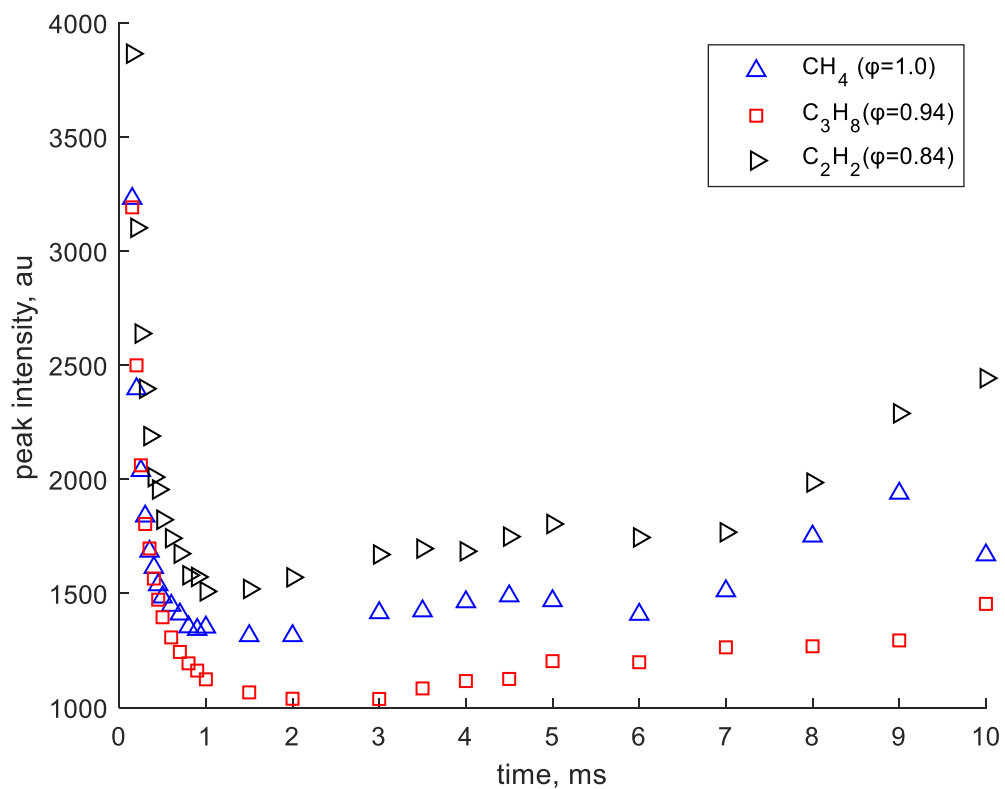


Figure 6.17: Peak OH* Intensity at 2226 K

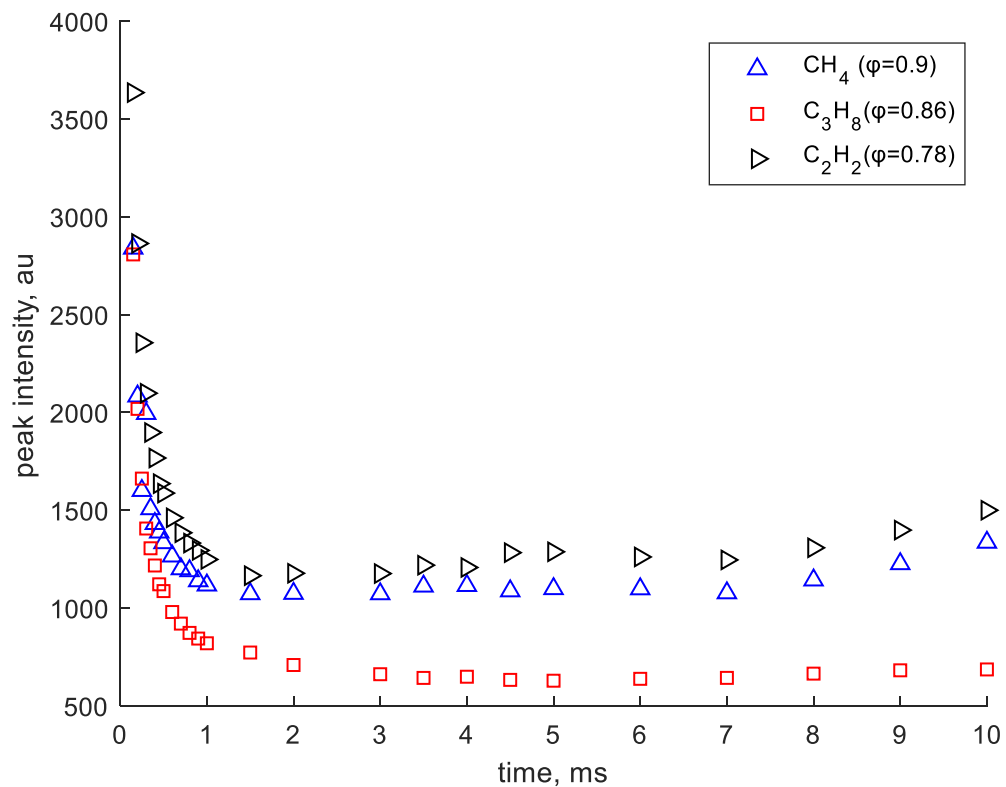


Figure 6.18: Peak OH* Intensity at 2134 K

Figure 6.19 - Figure 6.20 compares the spatially integrated OH* intensities obtained from the flame kernels of the three fuels at 2226 K and 2134 K respectively. Similar to the peak intensities, the relative order of magnitudes of integrated intensities is $C_2H_4 > CH_4 > C_3H_8$. Therefore, it can be inferred that the rate of heat release was highest in the ethylene flame kernel and lowest in the propane flame kernel. A second noticeable difference is the time it takes for the integrated OH* intensity values to drop to its minimum before rising again which had earlier been referred as the possible duration of chemical induction. According to Figure 6.19, the duration of chemical induction occurred in the following order: 200 μs in CH_4 , 300 μs in C_3H_8 and 500 μs in C_2H_4 . Although, chemical induction in propane ended much earlier than in ethylene, the radical generation rate is counterbalanced by losses resulting from rapid cooling of the overstretched kernel until 1 ms, hence the wider gap between the two. In the methane kernel the losses

is minimal, hence the closer gap between methane and ethylene despite having the initial value of integrated intensity being similar to the propane kernel at 150 μ s.

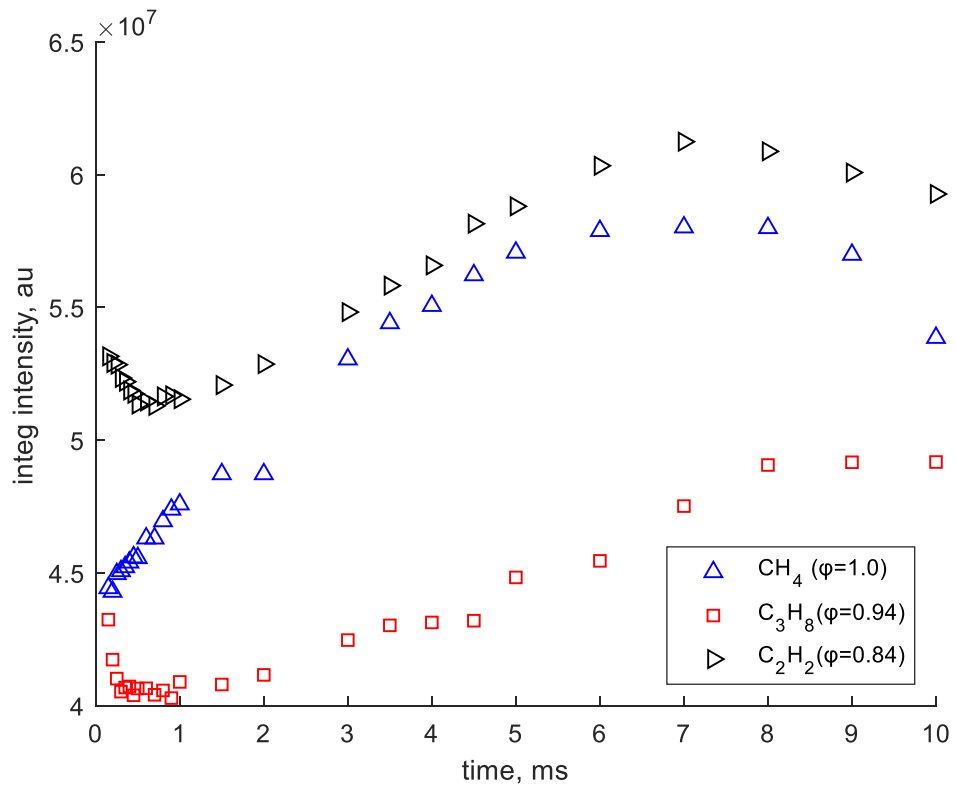


Figure 6.19: Integrated OH* Intensity at 2134 K

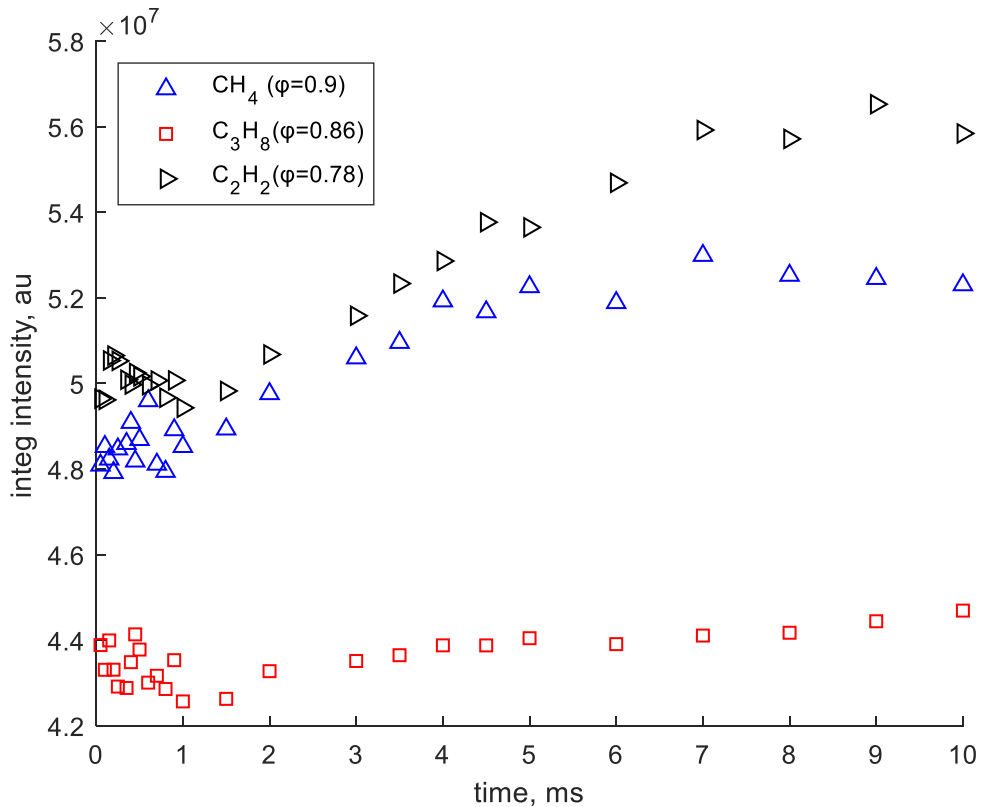


Figure 6.20: Integrated OH* Intensity at 2134 K

Figure 6.21 - Figure 6.22 compares the displacement of the flame front tip along the flow axis for the three fuel mixtures at 2226 K and 2134 K, respectively. As noted previously, the general trend in flame front displacement evolved from a decaying growth pattern in the 1 ms to a steady linear growth pattern onwards. Although flame kernels with the same adiabatic flame temperature were involved, clear differences can be seen beyond 1 ms when steady growth of the flame front is observed both upstream and downstream. Downstream, the propane kernel experienced the least growth while methane and ethylene grew at the same rate. Upstream the respective order in magnitudes was methane > ethylene > propane with maximum flame front displacement limits of 12.3, 9.9 and 6.7 mm respectively. The observed differences show the variation in stretch level of the upstream flame front due to the strain imposed by the flow. Also, the differences in growth of the flame kernels may be attributed to the differences in the mixture

Lewis Number (Le) values of the flow which in turn is inversely proportional to the unburnt mixture density. The Lewis number (Le) is defined as the ratio of thermal diffusivity (α) to mass diffusivity (D) and it's given by the equation [160]:

$$Le = \frac{\alpha}{D} = \frac{\lambda}{\rho_u * c_p * D_m} \quad 6.3$$

where: λ is the thermal conductivity, ρ_u is density of the unburnt mixture, c_p is the specific heat capacity at constant pressure, and D_m is the mixture-averaged diffusion coefficient.

Of the three mixtures (i.e., CH₄/air @ $\varphi=1.0$, C₃H₈/air @ $\varphi=0.94$ and C₂H₄/air @ $\varphi=0.84$) with AFT of 2226 K, methane is the only fuel lighter than air and the Le value of stoichiometric CH₄/air mixture is unity (i.e., $\varphi=1.0$, $Le=1$). In contrast, propane is the heaviest of the three fuels and the Le value of C₃H₈/air mixture ($\varphi=0.94$, $Le=1.4$) is the highest of the three. In [161], it was noted that mixtures with higher Le values would require a larger critical radius to be achieved for the flame to be sustained. Likewise, in the current investigation, the CH₄/air mixture with the least Le value would require lower **MIE** and less time to develop into a self-sustained flame kernel compared with the C₃H₈/air mixture. Hence, the order of duration in reaching a self-sustainable flame kernel is methane, ethylene, propane. A similar trend in axial FF distances is observed in leaner fuel/air mixtures (CH₄/air @ $\varphi=0.9$, C₃H₈/air @ $\varphi=0.86$ and C₂H₄/air @ $\varphi=0.78$) with $T_{ad}=2134$ K as shown by Figure 6.22. However, the differences become wider which is in accordance with their Le values. For fuels heavier than air (e.g., C₃H₈, C₂H₄) Le values increases with reduced φ values while the reverse is the case for fuels lighter than air (e.g., H₂, CH₄)[162].

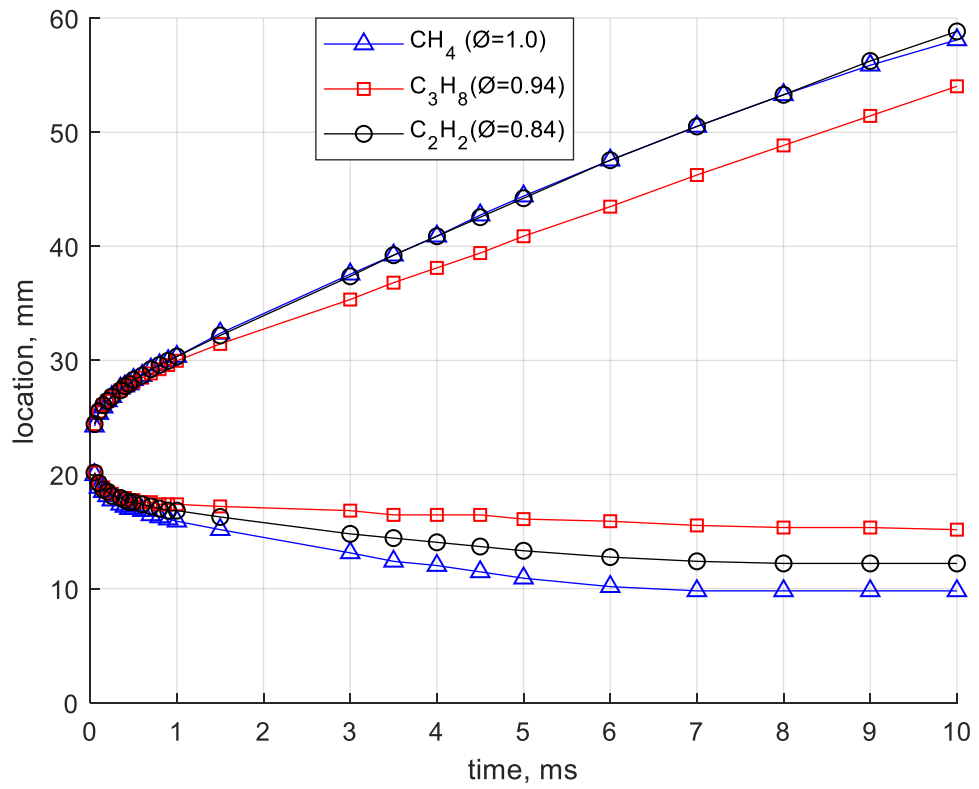


Figure 6.21: Axial Flame Front Locations at 2226 K

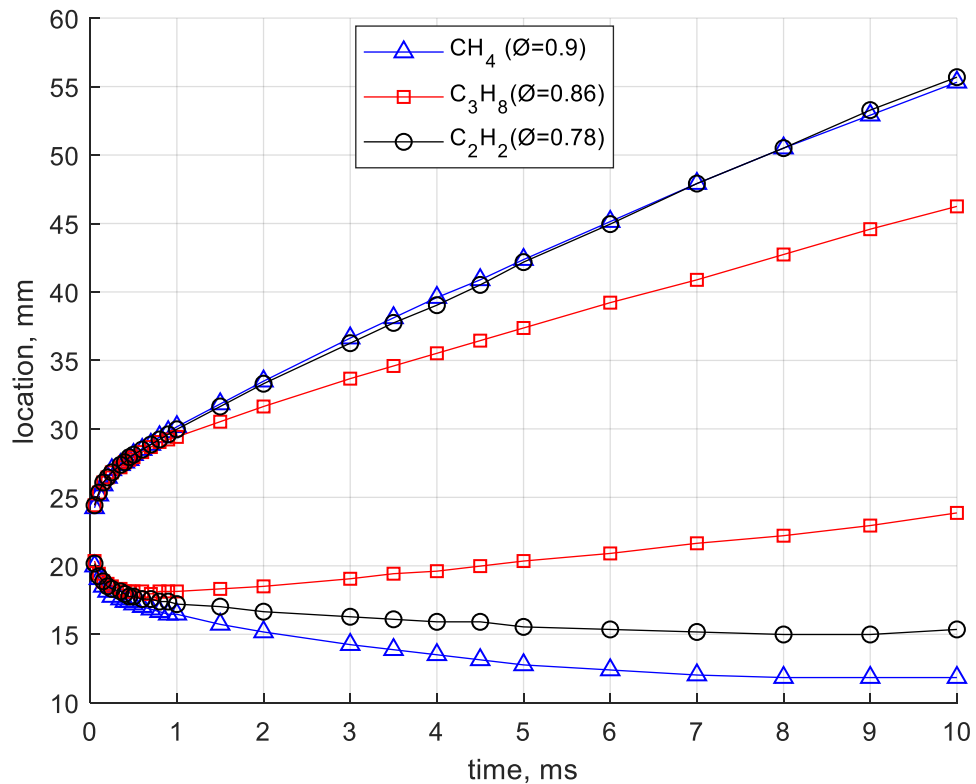


Figure 6.22: Axial Flame Front Locations at 2134 K

Figure 6.23 - Figure 6.24 compares the axial flame front propagation speed of the three mixtures at 2226 K and 2134 K adiabatic flame temperatures, respectively. As noted previously, the general trend for all the mixtures is that S_y decreases asymptotically from an initially stretched value to a steady value close to the unstretched laminar flame speed. However, obvious differences can be seen in the steady state propagation rates of each flame kernels. For example, at 2226 K flame temperature, the respective steady state S_{y+} values were 3.45, 3.45 and 2.49 m/s for methane, ethylene, and propane while the S_{y-} values were 1.35, 0.98 and 0.24 m/s respectively. Based on the S_{y-} comparison, it can be deduced that the propane mixture produced the slowest growing kernel while the methane mixture produced the fastest growing kernel despite having the same adiabatic flame temperature. This is unexpected because ethylene has the highest estimated laminar flame speed (i.e., $S_L = 3.87$ m/s for C_2H_4/air) while methane has the least value (i.e., $S_L = 3.06$ for CH_4/air). By igniting leaner mixtures with reduced flame temperature of 2134 K, a similar trend is repeated with the respective S_{y-} values

of methane, ethylene and propane reduced by 32, 62 and 329% respectively. This behaviour reaffirms the earlier assertion that flame kernels of the different fuels may be stretched differently by the same amount of flow-imposed strain despite having the same flame temperature and the stretch may increase at lower flame temperatures.

To better illustrate the relative differences in growth rate, the axial flame front propagation speed is normalised by the unstretched laminar flame speed. Figure 6.25 - Figure 6.26 compares the normalised propagation speed of the axial flame front for the three fuel blends with adiabatic flame temperatures of 2226 K and 2134 K respectively. The first observation from the results is that as the kernels grew in size, the propagation rate of the downstream flame front decreases until it converges to a value close to the unstretched laminar value, S_L . In contrast, the propagation rate of the upstream flame front continues to decrease after reaching the unstretched value S_L before converging to its asymptotic values. This is shown by the fact that the normalised S_{y-} values lie farther above the -1.0 line unlike the normalised S_{y+} which lie somewhat around the 1.0 line. This behaviour indicates a negatively stretched flame front which could be due to several factors. One of the factors noted previously is that the flame front is impacted by flow-induced strain. The impact flow-induced strain could be in the form of wrinkling which could lead to differences in the surface area between the upper and lower flame front [163]. It could also be that the local S_L values have changed due to varying compression of the mixture in both the upstream and downstream region by the expanding flame kernel [90]. Another factor responsible for different stretching of the upper and lower flame fronts is the different curvature of the flame surface. Overall, the stretch increased when the mixtures were leaned (i.e., lowered flame temperature). A second feature observed from the results is the differences in stretch sensitivity of the flame kernels due to different fuel composition despite having the same flame temperature. For example, at 2226 K flame temperature, S_{y+} variation from S_L were +8.7%, -13.9% and -28.5% for methane, ethylene, and propane respectively while the respective S_{y-} variation from S_L were -64%, -81% and -100%. Following a reduction in flame temperature from 2226 K to 2134 K, wider differences were observed. For example, at 2134 K flame temperature, S_{y+}

variation from S_L were -4%, -24% and -49% for methane, ethylene, and propane respectively while the respective S_{y-} variation from S_L were -76%, -91% and -120%. Therefore, it can be deduced that propane kernel is the most sensitive to stretch while methane kernel is the least sensitive to stretch. The measure of flame sensitivity to stretch is given by the Markstein Length (ℓ) which in turn depends on the Effective Lewis Number, Le of the mixture. This is shown by equation 6.4, in which the stretched flame speed (S_f) is expressed as the difference between unstretched laminar value, S_L and the product of Markstein Length, ℓ and the stretch rate, K [162].

$$S_f = S_L - \ell * K \quad 6.4$$

Just like Le , the variation of ℓ with mixture composition widens as the flow becomes leaner. The reason is that in the lean range, Le increases for fuels heavier than air (e.g., C_3H_8 , C_2H_4) and decreases for fuels lighter than air (e.g., H_2 , CH_4). By comparing both plots, it is obvious the variation of S_{y-} from S_L (or the stretch level) is widened by a decrease in mixture AFT from 2226 K to 2134 K.

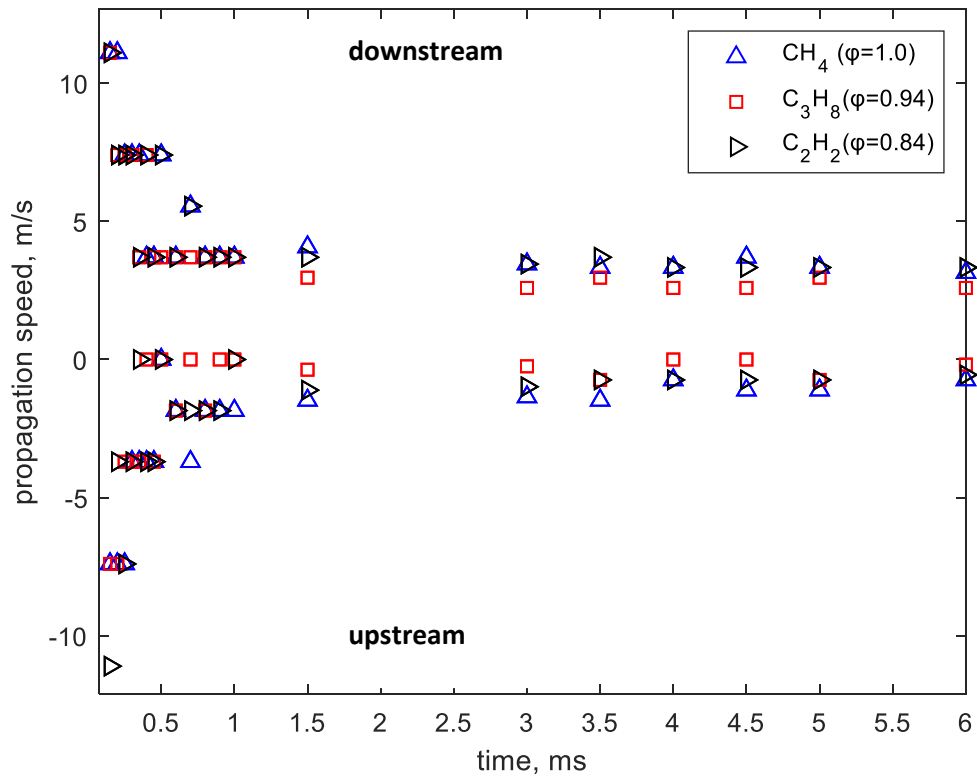


Figure 6.23: Axial flame front propagation rate at 2226 K

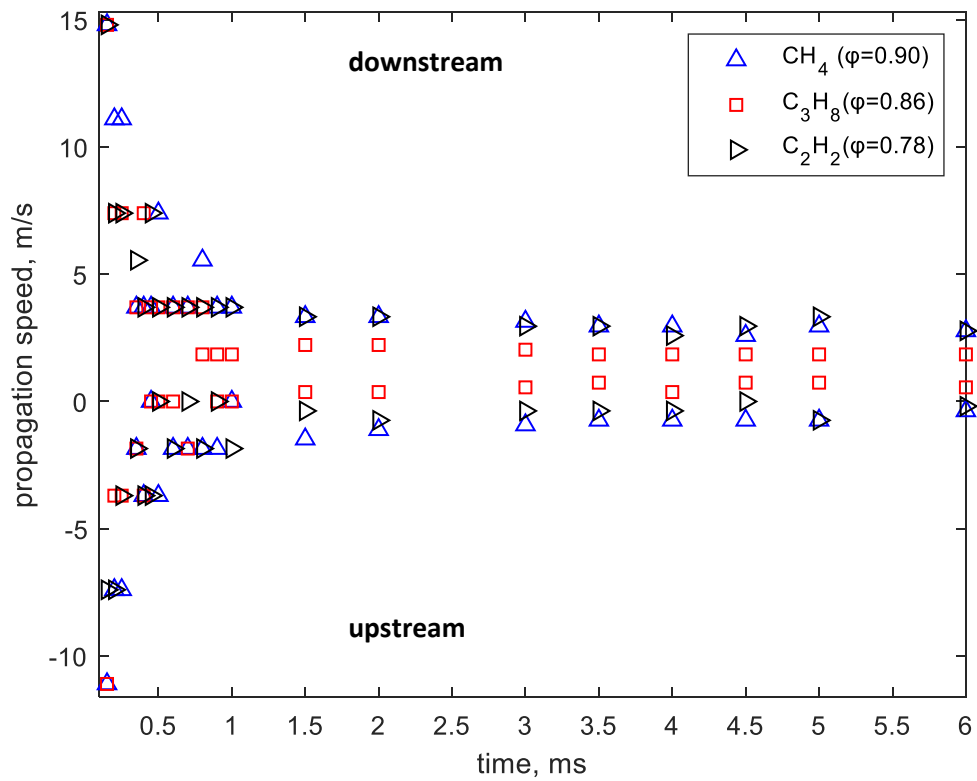


Figure 6.24: Axial flame front propagation rate at 2134 K

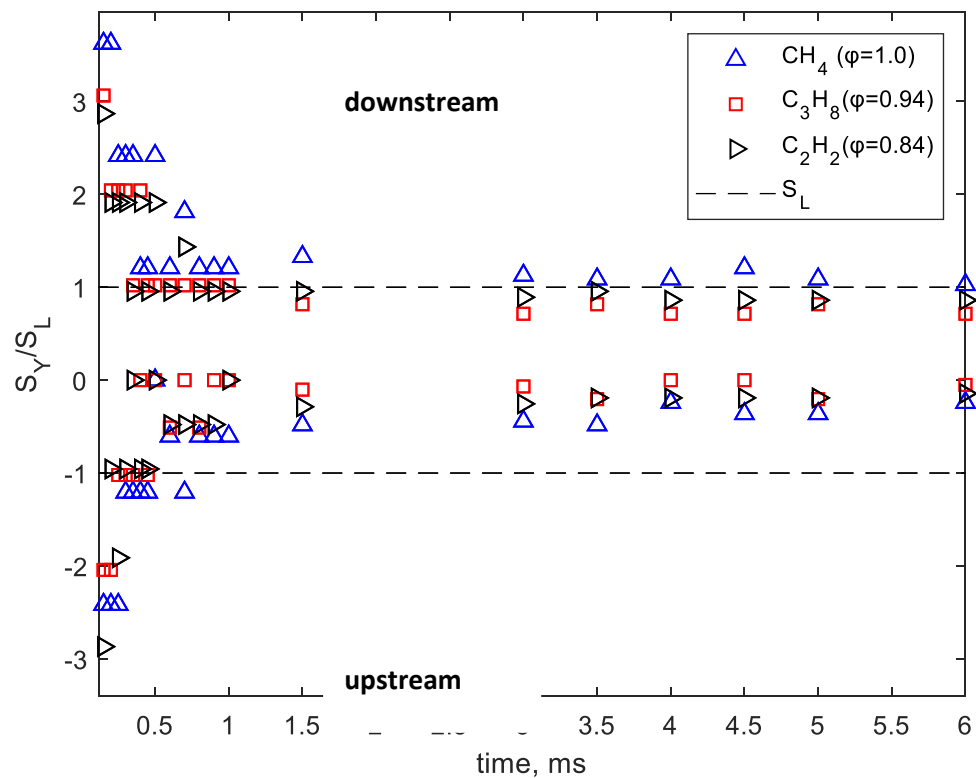


Figure 6.25: Normalised flame propagation rate at 2226 K

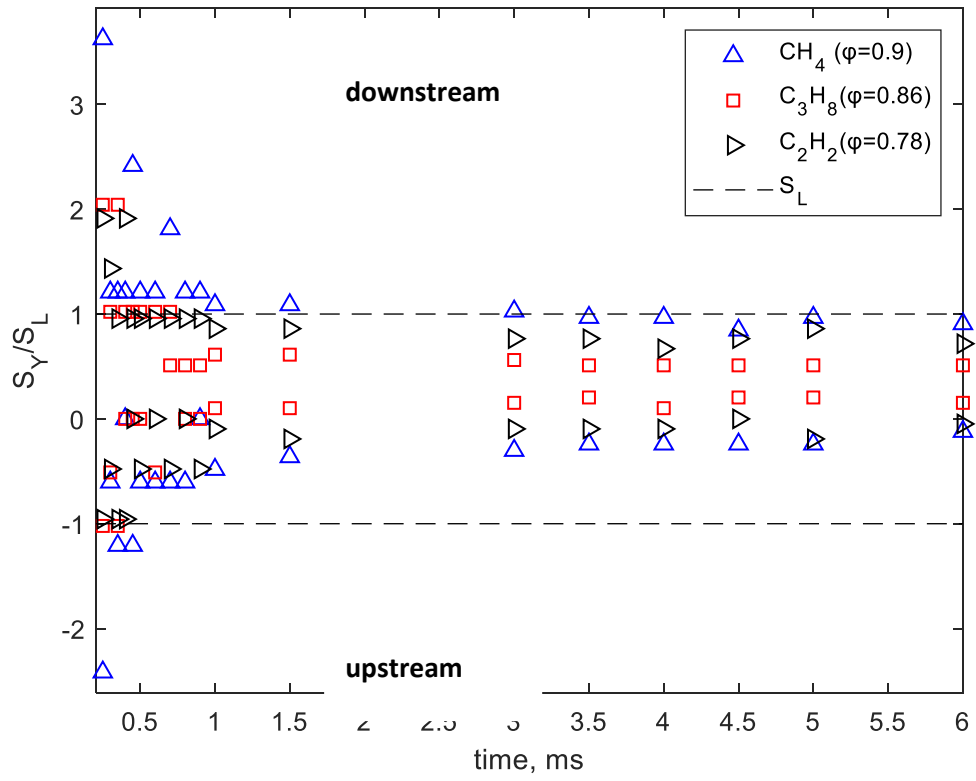


Figure 6.26: Normalised flame propagation rate at 2134 K

6.4 Fuel effects based on constant unstretched laminar flame velocity.

To further establish the influence of fuel properties on the propagation of the laser-ignited flame kernel, flow mixtures with different chemical composition are varied in such a way that a constant laminar flame velocity (LFV) is maintained. In this section, the effect of the fuel on the characteristics of the flame kernel is reported for three gaseous fuel/air mixtures (CH_4/air , $\text{C}_3\text{H}_8/\text{air}$, and $\text{C}_2\text{H}_4/\text{air}$) with the same LFV. At first, OH^* chemiluminescence images of the flame kernels of three laminar mixtures (i.e., CH_4/air @ $\phi=1.0$, $\text{C}_3\text{H}_8/\text{air}$ @ $\phi=0.92$ and $\text{C}_2\text{H}_4/\text{air}$ @ $\phi=0.74$) with a LFV of 41 m/s were obtained and their characteristics compared to quantify the differences due to fuel type. This was followed by further investigation of the characteristics under leaner mixtures (i.e., CH_4/air @ $\phi=0.9$, $\text{C}_3\text{H}_8/\text{air}$ @ $\phi=0.84$ and $\text{C}_2\text{H}_4/\text{air}$ @ $\phi=0.7$) with SL value of 29 cm/s. Overall six (6) flow conditions were investigated which are given in table 6.3. From the table, it is evident that mixtures with the same LFV may have varying AFT. A graph showing the selected mixture composition in the S_L vs ϕ curve is given in Figure 6.27. Besides S_L being constant, the mixtures were selected such that all experimental conditions including the bulk flow velocity were kept constant.

Table 6.3: Summary of selected Laminar Flow conditions based on constant S_L .

Test Case	ϕ	Flow Rate (lpm)				U_0 (m/s)	Re (Pa.s)	T_{ad} (K)	u_L (cm/s)	$\frac{\rho_u}{\rho_b}$
		Air	CH_4	C_3H_8	C_2H_4					
Laminar mixture @ 41 m/s	1	23.8	2.5	0	0	1.15	1694	2226	41	7.47
	0.92	23.8	0	1	0	1.15	1839	2196		8.60
	0.74	25.9	0	0	0.9	1.18	1978	2062		6.92
Laminar mixture @ 29 m/s	0.8	25.0	2.1	0	0	1.19	1750	1966	29	6.60
	0.76	25.0	0	0.85	0	1.19	1865	1976		7.74
	0.64	26.6	0	0	0.75	1.20	1894	1882		6.31

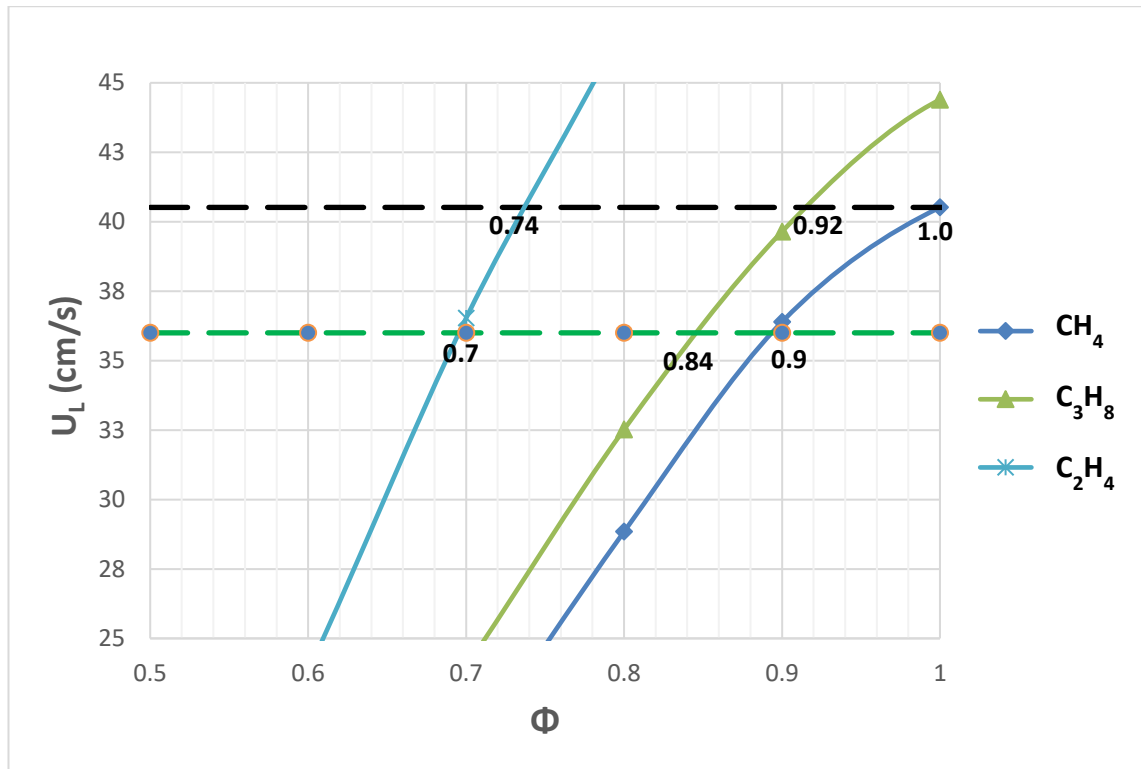


Figure 6.27: Selected flow conditions on LFV – ϕ plots

Figure 6.28 are sequences of mean OH* chemiluminescence images which visualizes the comparative characteristics of the flame kernels developing in different laminar fuel/air mixtures (i.e., CH₄/air @ $\phi=1.0$, C₃H₈/air @ $\phi=0.92$ and C₂H₄/air @ $\phi=0.74$) with a constant LFV of 41 cm/s. As expected, all the flame kernels exhibit common geometric features in their development (i.e., evolved from a two-lobe symmetric toroid to a three-lobe asymmetry and finally to a flame ball). However, there are noticeable differences in their growth especially for the propane mixture which produced the smallest kernel. From the pictures, it is seen that in the first 400 μ s, the kernels initially grow at the same rate in all directions, however from 500 μ s to 2 ms, the propane kernel experienced more elongation of the front lobe which in turn affected the overall growth. On the other hand, both methane and ethylene kernels grew at nearly the same rate producing bigger flame kernels than propane. Comparing the shape of the upstream flame front in late kernels, it is obvious the difference in growth rate is mostly influenced by flow-induced strain which is expected to be highlighted through analysis of the images.

In addition, the reactivity and strength of each kernel would be highlighted through analysis of the OH* intensities.

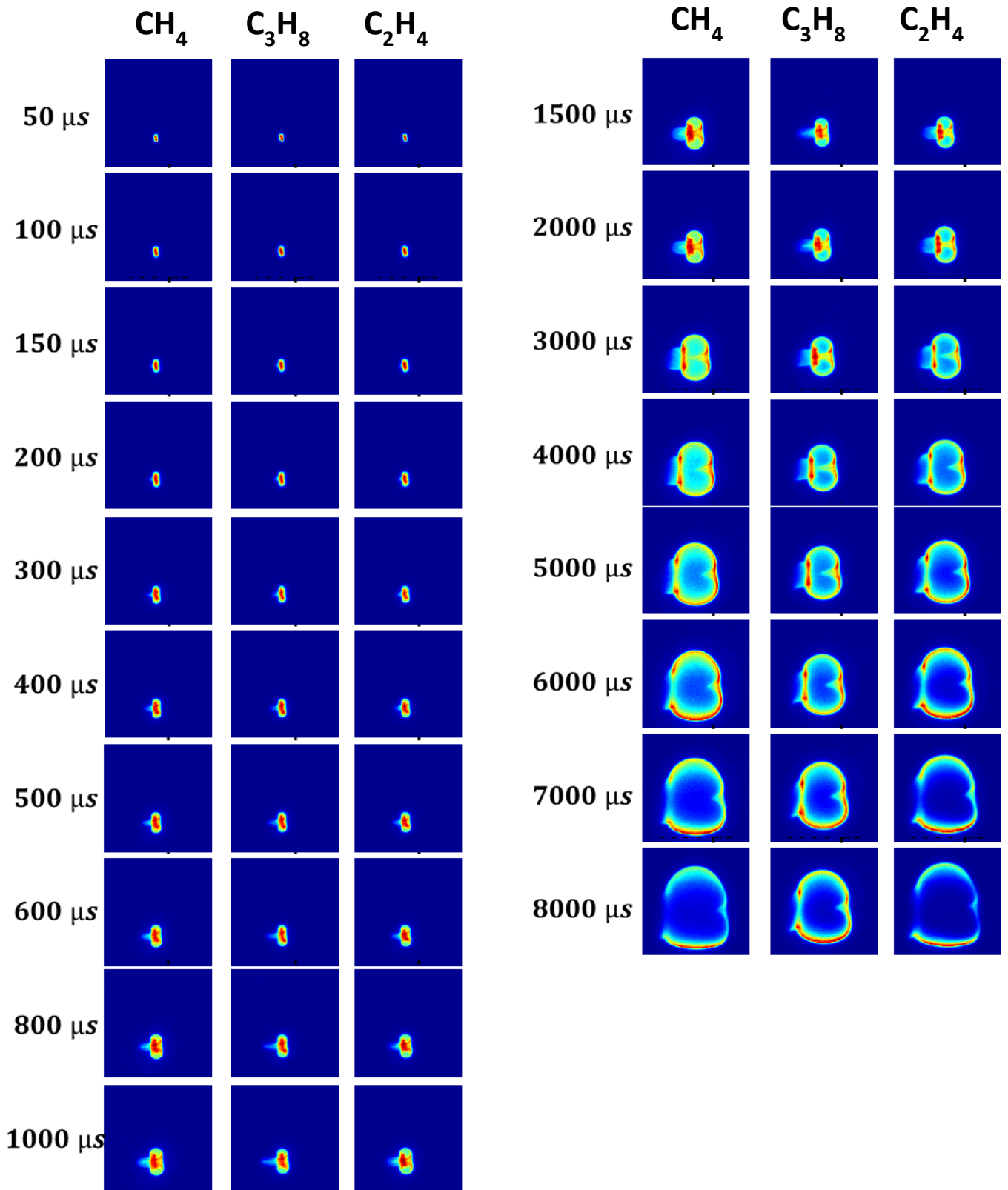


Figure 6.28: Visualization of fuel composition effects of fuels at constant LFV (Image FOV: 56x56 mm²)

Figure 6.29 - Figure 6.30 compares the peak OH* intensities obtained from the flame kernels of the three fuels at 41 cm/s and 29 m/s unstretched laminar flame velocities, respectively. By comparing both results, it appears the peak OH* intensities followed the similar trend as it did when a constant flame temperature was maintained. For example, in the mixtures with LFV of 41 cm/s, the peak OH* intensities seem to reveal that the most reacting mixture was ethylene, while propane and methane had similar reactivity. However, a closer look at the decay from the initial peak OH* intensity, it is evident that the propane kernel decayed much faster than the rest which explains the smaller size of at the developed stage. In the leaner mixtures with LFV of 29 cm/s, both methane and ethylene kernels produced a higher steady peak OH* intensity of 800 AU at 2 ms following the initial decay while the propane kernel produced the lowest steady peak OH* intensity of 500 AU at 3.5 ms following an initial decay. This trend in reactivity is unexpected by considering the flame temperatures estimated. As shown in Table 6.3, the order of reactivity with respect to the AFT should be methane>propane>ethylene for the in the mixtures with LFV of 41 cm/s while that of the lean mixtures (with LFV of 29 cm/s) should be propane>methane>propane. This behaviour may be linked to the fact that the actual flame temperature deviates from the AFT in a proportion similar to the stretching of the flame front [162]. The stretch effect in turn depends on Lewis number of the deficient component (i.e., the Fuel) as noted previously.

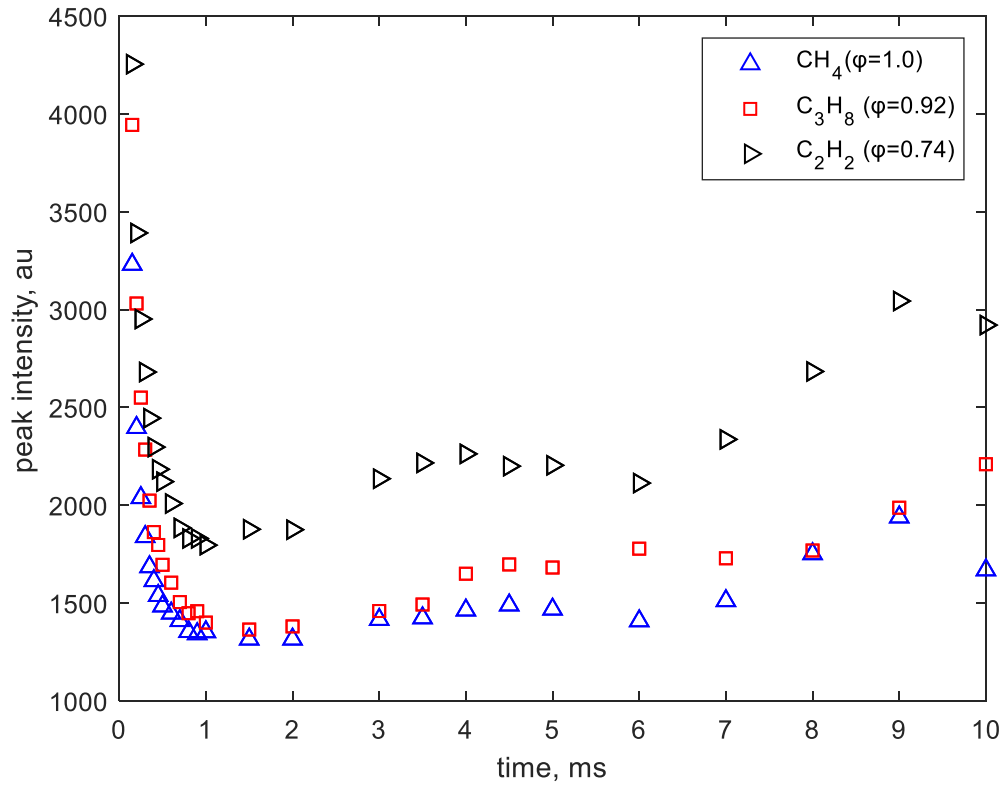


Figure 6.29: Peak OH* Intensity at 41 cm/s

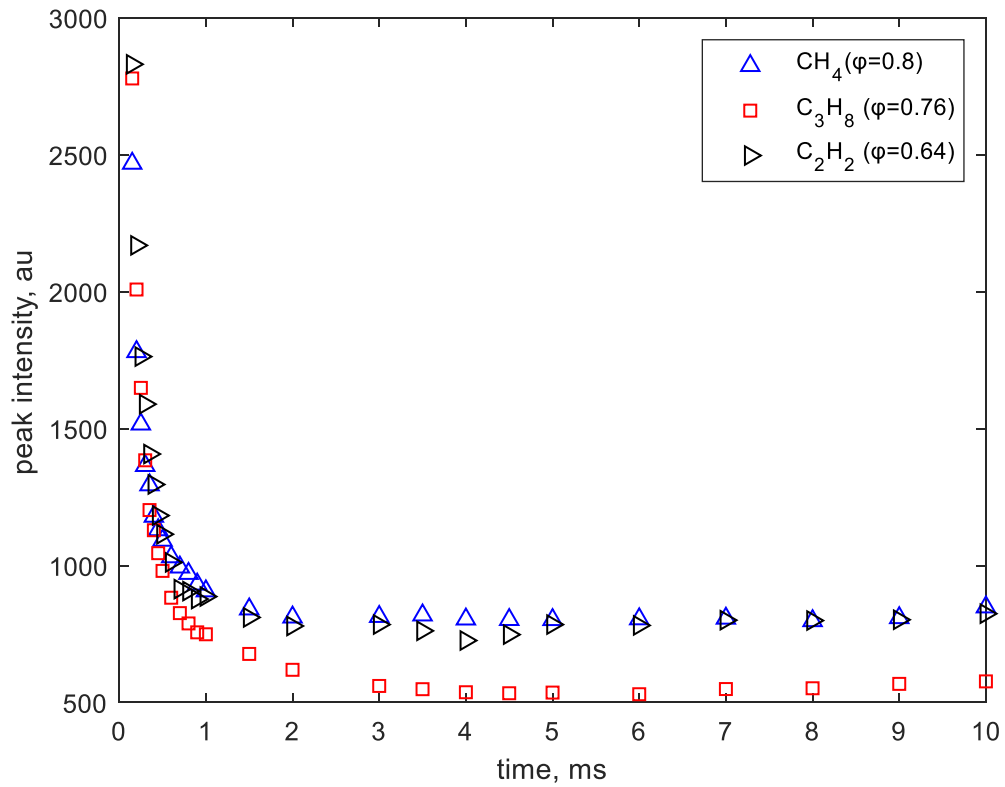


Figure 6.30: Peak OH* Intensity at 29 cm/s

Figure 6.31 - Figure 6.32 compares the spatially integrated OH* intensities obtained from the flame kernels of the three fuels with LFV of 41 cm/s and 29 cm/s, respectively. From the results, the magnitudes of integrated intensities occur in the following order: $C_2H_4 > CH_4 > C_3H_8$. Therefore, it can be inferred that under the given flow conditions the rate of heat release was highest in the ethylene flame kernel and lowest in the propane flame kernel. A second noticeable difference is the time it takes for the integrated OH* intensity values to drop to its minimum before rising again which had earlier been referred as the possible duration of chemical induction. For the mixtures with LFV of 41 m/s, the duration of chemical induction occurred in the following order: 200 μs in CH_4 , 400 μs in C_3H_8 and 250 μs in C_2H_4 . Comparing the kernel development in the two Figures shows that at lower LFV, more time was required to see a rise in integrated OH* intensity. Also, the late kernels were marked by wider difference in integrated OH* intensity for the leaner mixtures.

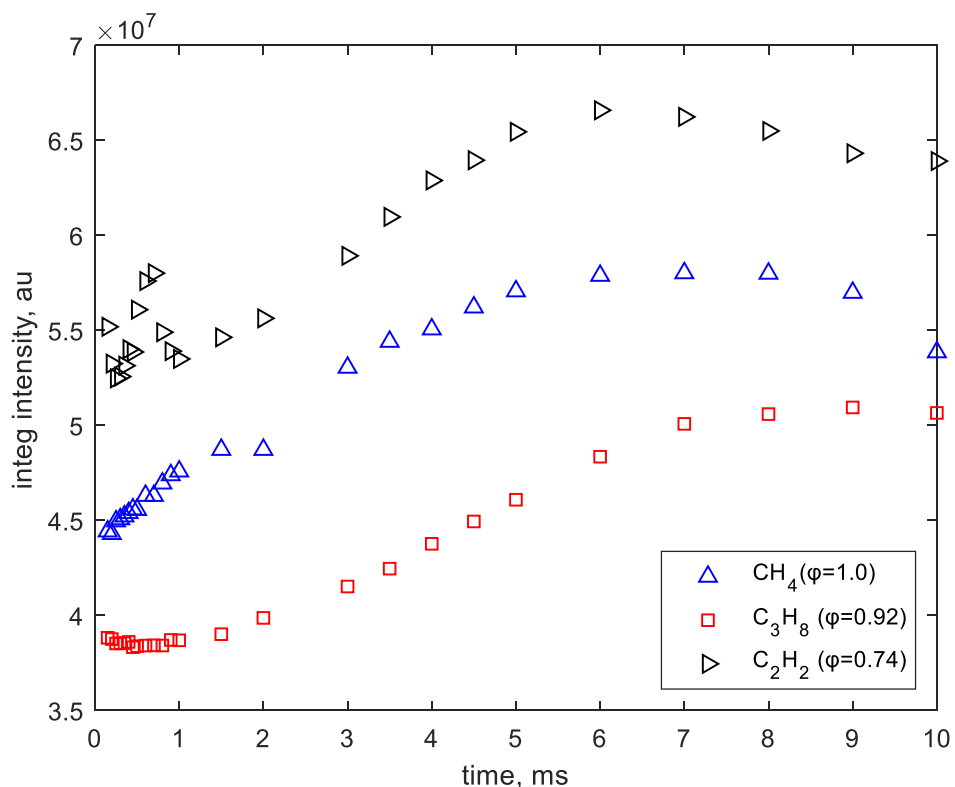


Figure 6.31: Integrated OH* Intensity at 41 cm/s

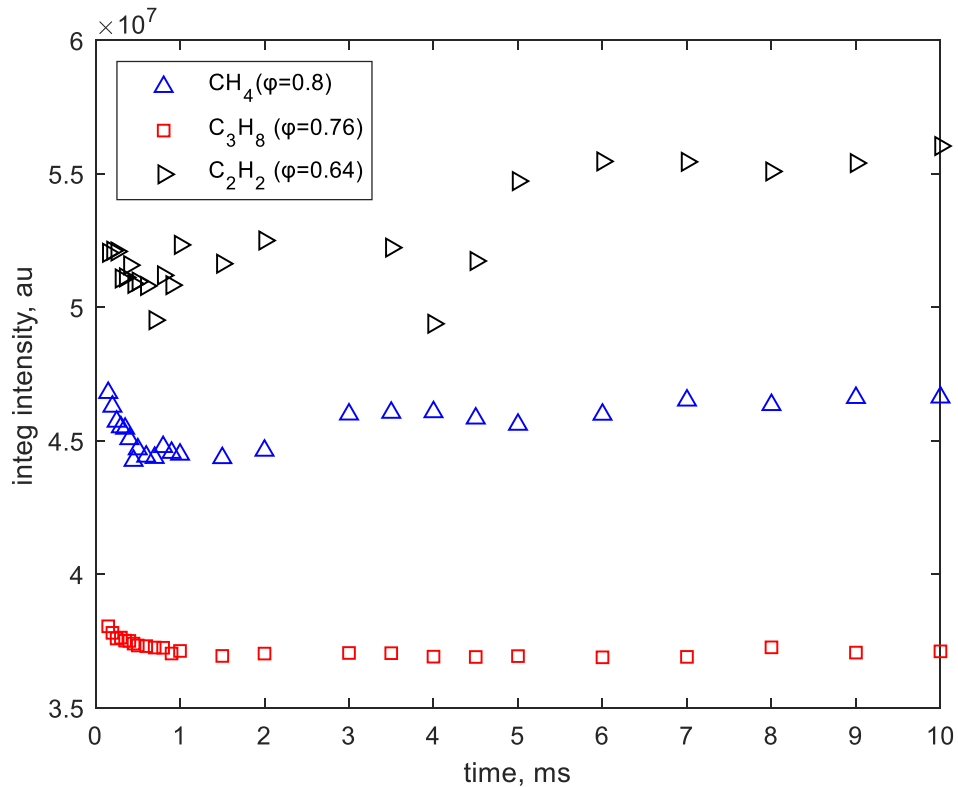


Figure 6.32: Integrated OH* Intensity at 29 cm/s

Figure 6.33 - Figure 6.34 compares the displacement of the flame front tip along the flow axis for the three fuel mixtures with at LFV of 41 cm/s and 29 cm/s, respectively. As noted previously, the general trend in flame front displacement evolved from a decaying growth pattern in the 1 ms to a steady linear growth pattern onwards. The general trend in all the graph is a symmetric axial growth with propane mixture having the slowest growth. Under the first condition, the influence of the fuel composition is clearly shown by the distances travelled in each kernel in both the upstream and downstream direction. For example, under mixture conditions with LFV of 41 cm/s, the respective Y_+ values at 7 ms delay was 31, 29 and 33 mm in methane, propane, and ethylene. Although, there is hardly a noticeable difference in the kernel height between CH₄/air and C₂H₄/air mixtures; the height of the propane kernel clearly supports the fact that higher Le values leads to slower growth in mixtures heavier than air. For the second condition in which the three mixtures are further leaned with a lower LFV of 29 cm/s, a remarkable difference is seen in the growth of each kernel in both the upstream and downstream directions. This is also in line with the variation of mixture

effective Le values which in the lean flammability range increase for heavier than air (e.g., C_3H_8 , C_2H_4) but decreases for fuels lighter than air (e.g., H_2 , CH_4).

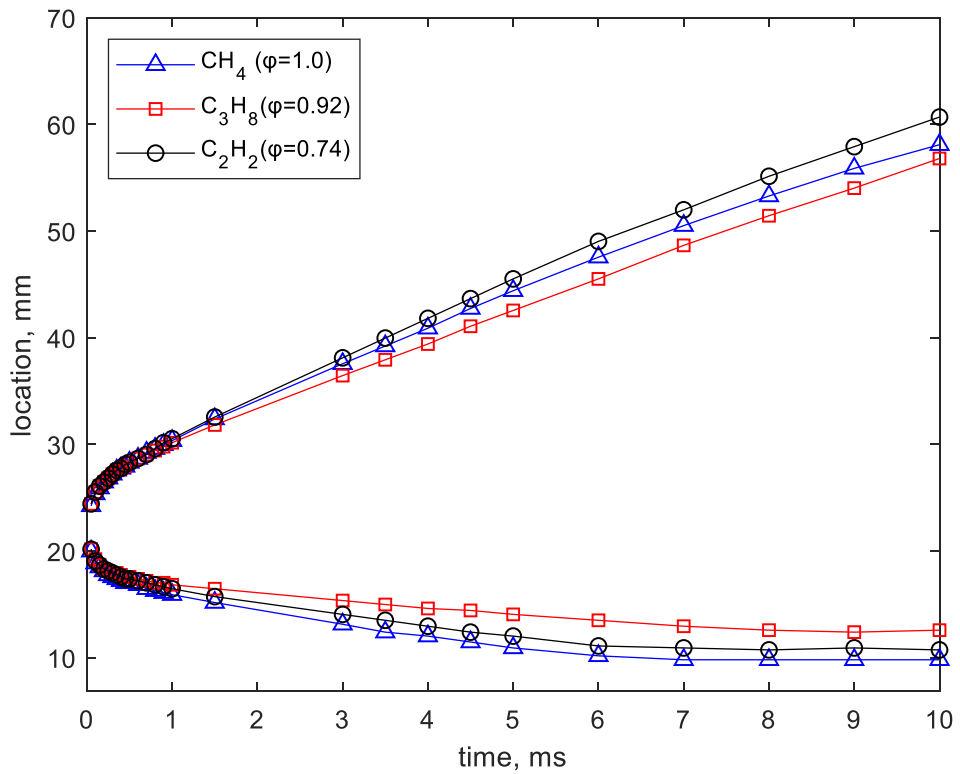


Figure 6.33: Axial flame tip locations at 41 cm/s

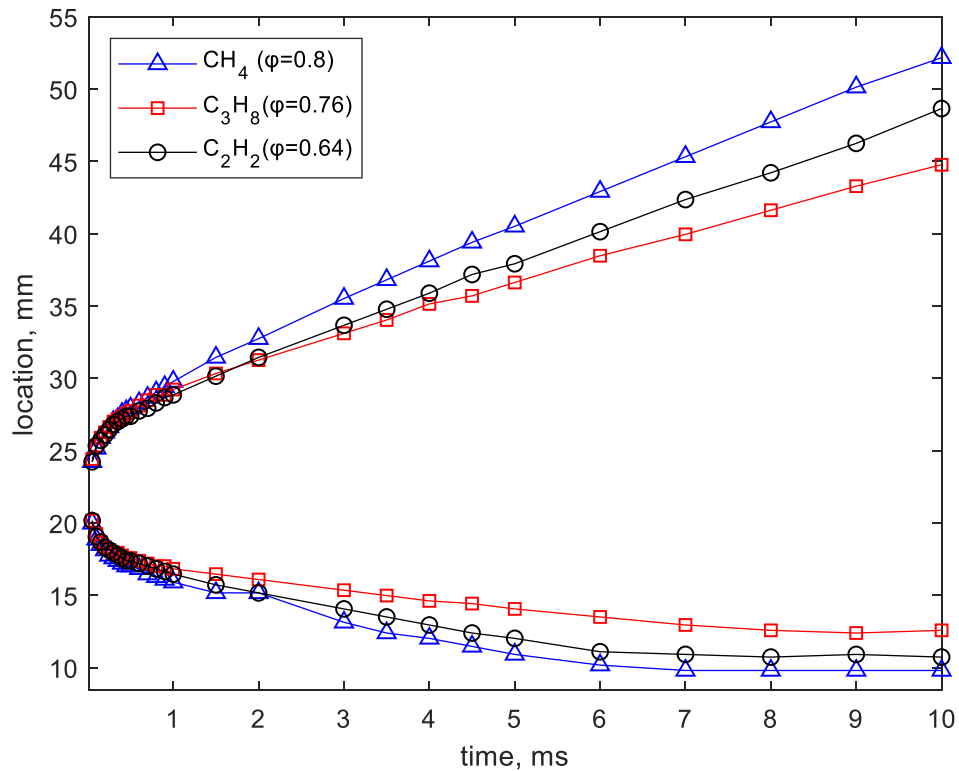


Figure 6.34: Axial flame tip locations at 29 cm/s

Figure 6.35 - Figure 6.36 compares the axial flame front propagation speed of the three mixtures with at LFV of 41 cm/s and 29 cm/s, respectively. As noted previously, the general trend for all the kernels is that S_{y+} decreases asymptotically from an initially stretched value to a steady value close to the unstretched laminar flame speed. However, obvious differences can be seen in the steady state values of the propagation rates of each flame kernel with the propane kernel being the slowest while the ethylene kernel was the fastest. For example, with mixture LFV of 41 cm/s, the steady state S_{y+} values were 3.33 m/s in methane (a 9% increase in S_L), 3.70 m/s in ethylene (a 30% increase in S_L), and 2.96 m/s in propane (a 16% decrease in S_L). Based on the S_{y+} comparison, it can be deduced that the propagation rate of the kernels did not follow the correct order expected based on the S_L values which is 3.06 m/s for methane, 2.84 m/s for ethylene, and 3.52 m/s for propane. It is however in agreement with the earlier observed trend in heat release rate which is indicated by the integrated OH* intensity. Further leaning the mixtures resulted in a slightly different outcome in which the fastest growing kernel was that of methane while the propane kernel remained the slowest. For

example, at LFV of 29 cm/s, the steady state S_{y+} values were 2.59 m/s in methane (a 35% increase in S_L), 2.22 m/s in ethylene (a 21% increase in S_L), and 1.85 m/s in propane (a 17% decrease in S_L) when the unstretched laminar values were 1.91, 1.83 and 2.24 m/s respectively. In this result, the measured flame front propagation rate is not in agreement with the estimated unstretched laminar flame velocity of the flame and the observed trend is not in agreement with the integrated OH* intensity. This behaviour reaffirms the earlier assertion that flame kernels were stretched differently depending on the fuel.

To better illustrate the relative differences in growth rate, the axial flame front propagation speed is normalised by the unstretched laminar flame speed. Figure 6.37 - Figure 6.38 compares the normalised propagation speed of the axial flame front for the three fuel blends with mixture LFV of 41 cm/s and 29 cm/s, respectively. Analysis of the downstream flame front shows that the propagation rate, S_{y+} decreased from an initially stretched value until it converges to a value close to the unstretched laminar value, S_L . The ethylene mixture was the first to reach a stable value $1.3S_L$ at 3 ms, followed by methane (i.e., $1.09S_L$ at 3.5 ms) and finally propane (i.e., $0.84S_L$ at 3.5 ms) which was the least stretched. In the leaner mixtures at 29 cm/s, methane became more stretched than ethylene (i.e., $1.35S_L$: $1.21S_L$) while propane remained the least stretched (i.e., $0.83S_L$). Analysis of the upstream flame front shows that the propagation rate, S_{y-} continues to decrease after reaching the unstretched value S_L before converging to its asymptotic values. The difference between the two flame fronts is that while it is positively stretched downstream, the flame front upstream is negatively stretched. This explains the rationale for S_{y+} lying above the 1.0 line and for S_{y-} lying above the -1.0 line. As previously explained, the difference may be due to the variations in surface area for the flame front as well as the varied compression effect of flame expansion on flow ahead of the flame which could be revealed through further analysis. With respect to the stretch sensitivity of the flame kernel, the results show that the ethylene fuel had the most influence while propane had the least influence for mixtures with LFV of 41 cm/s. However, at a lower LFV of 29 cm/s, methane was the most influential while propane had the least influence. As suggested earlier, this behaviour may be due to a higher variation of C₂H₄/air flame temperature

from the AFT which can be linked to stretch dependence on Lewis number of the deficient component (i.e., the Fuel).

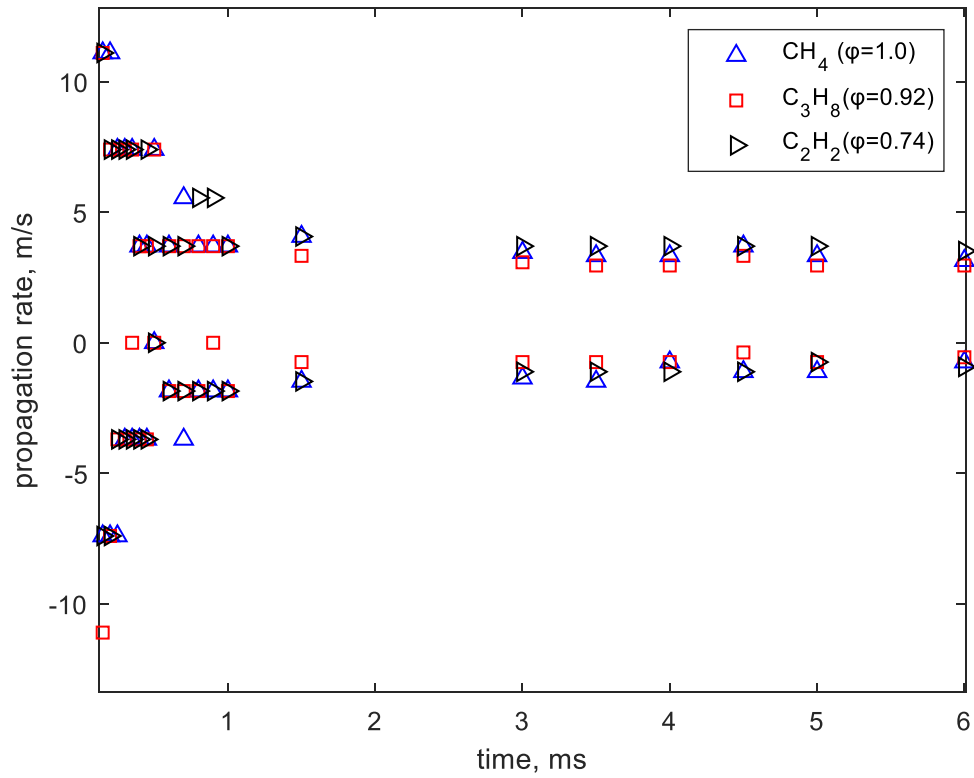


Figure 6.35: Axial propagation rate at 41 cm/s

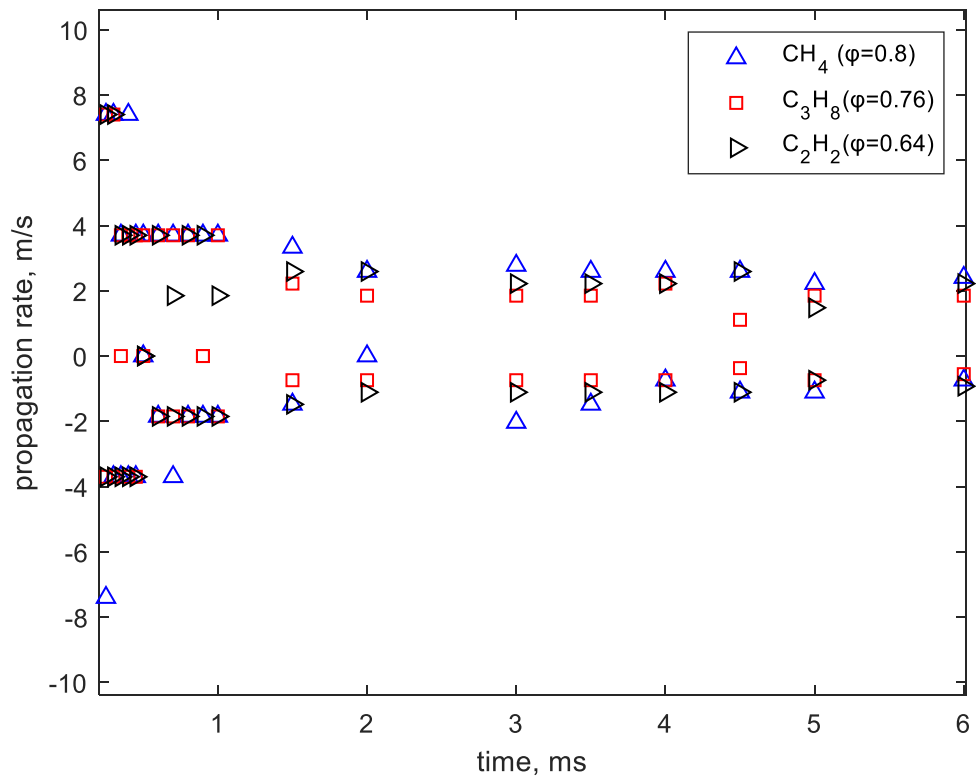


Figure 6.36: Axial propagation rate at 29 cm/s

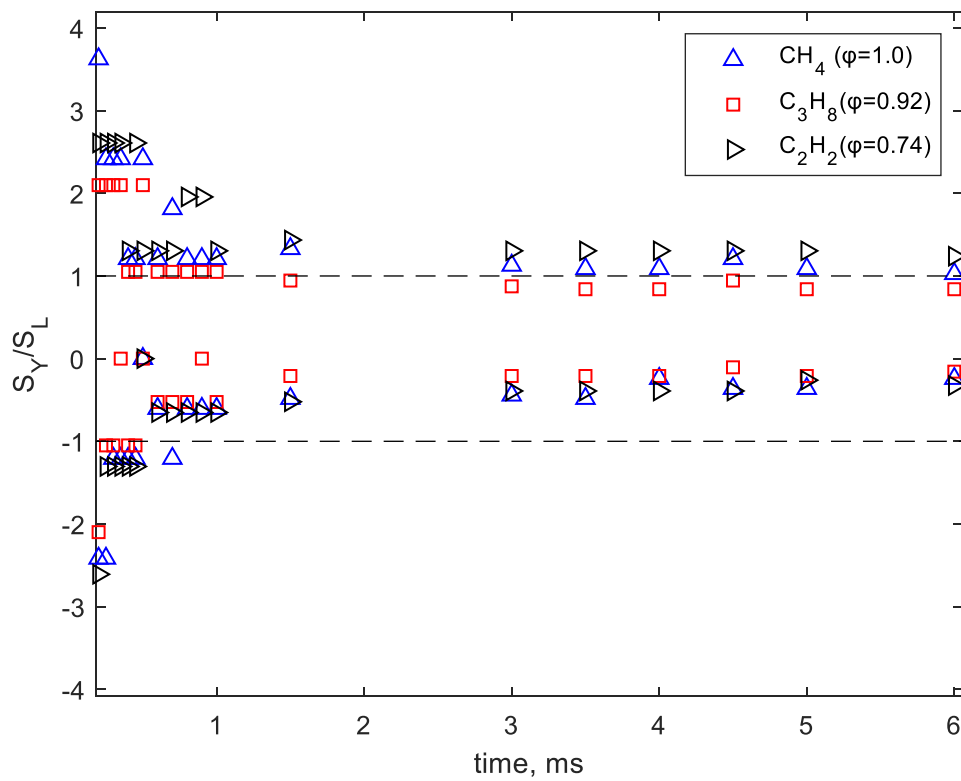


Figure 6.37: S_{y+}/S_L at 41 cm/s

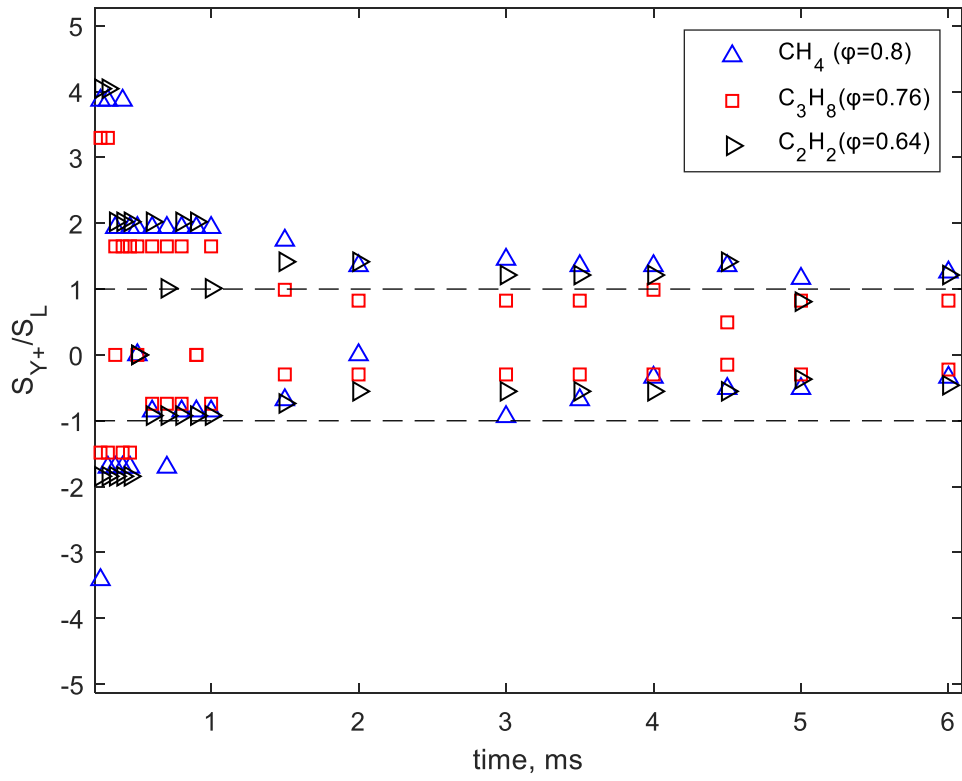


Figure 6.38: S_{y^+}/S_L at 29 cm/s

6.5 Conclusions

The effect of the fuel properties on the development characteristics of the LISI flame kernel has been investigated through analysis of the mean OH* chemiluminescence images. First, the flame kernel development was compared at changing equivalence ratios while maintaining a constant flow velocity in both laminar and turbulent flow environment. Also, a second and third comparison was made at constant AFT and LFV, respectively to investigate the sensitivity to the selected fuel mixture. All the experiments were performed by repeated laser ignition of different Fuel/Air mixtures (CH₄/Air, C₃H₈/Air and C₂H₄/Air) in a co-flow burner made of smoothly contoured nozzle with contraction and the acquired images included both kernel development and initial propagation covering a temporal range of 0.05 – 10 ms. A summary of the findings based on both qualitative visualization and quantitative analysis are as follows:

Effects of variation in Mixture Equivalence Ratio

1. Visualisation observation of the kernel development in lean and stoichiometric mixtures (i.e., $\varphi=1.0$ to $\varphi=0.8$) produced similar features, however there were noticeable differences in the size and reactivity of the developing flame kernels.
2. The peak OH* intensities showed that a unit reduction in φ produced a similar reduction in the kernel peak OH* intensity during its development. This indicates more heat losses in kernels at lower φ values which could hinder its development to a stable flame kernel. At the developed kernel stage, the peak intensities increased for the near stoichiometric mixture kernels but remained constant at $\varphi=0.8$ which means lower heat release rates.
3. Also, the integrated OH* intensities showed that lower equivalence ratios produced both weaker kernels and increased chemical induction duration.
4. the kernel growth is slower when lean mixtures (i.e., $\varphi = 0.9, 0.8$) are compared with the stoichiometric mixture. Analysis showed the leanest flame

- kernel required 9 ms to travel 28 mm upstream compared to 7 ms required by the stoichiometric flame.
5. The tip displacement of the flame front in laminar flow mixtures, reveals a nearly symmetric growth curve in which the distance travelled decreased from $\varphi=1.0$ to $\varphi=0.8$. Analysis shows the leanest flame kernel required 9 ms to travel 28 mm upstream compared to 7 ms required by the stoichiometric flame. In the turbulent flow mixtures, the distance travelled increased in a non-symmetric manner which led to reduction in the gap between the flame fronts.
 6. For all φ values, the general trend shows that the flame front growth rate during kernel development decreases asymptotically from a highly stretched value to a value close to S_L after which a nearly constant speed is maintained. During development, the mixture with the least φ value experienced the most stretch. In the developed kernels, the steady state values of S_{y+} increased in order of the equivalence ratios (from $\varphi=0.8$ to $\varphi=1.0$).
 7. Further sensitivity analysis reveals that the decreasing trend in all parameters moving from $\varphi=1.0$ to $\varphi=0.8$ follows a linear relationship for laminar flows. This, however, was not the case in turbulent flows.

Effects of variation in Fuels with constant AFT

1. For selected mixtures of 2226 K AFT (i.e., methane/air, propane/air and ethylene/air), propane produced the slowest growing flame kernel while methane and ethylene kernels were growing at the nearly the same rate.
2. Analysis of the peak OH^* intensities showed that the slower growth in the propane kernel was due to faster cooling. This could be observed in the excessive stretch and local quenching at the front lobe. The peak intensities of the developed kernels also showed that the lowest heat release rates occurred in propane/air and highest in the ethylene/air.
3. The spatially integrated OH^* intensity indicate that the ethylene/air mixture produced the strongest flame kernel while propane/air mixture produced the weakest flame kernel. This also showed that the duration of chemical induction was shorter in methane/air mixture than the rest.

4. Analysis of the axial flame front locations showed that the upstream propagation (Y_*) differed for all the three mixtures. The order of magnitude of Y_* was methane > ethylene > propane which could be linked to the mixture Lewis Number or Density. This affected the expected symmetry in axial growth curve of laminar flame kernel.
5. From the S_{y*} plots, the general trend showed that the propagation rate of the flame front decreases asymptotically from a highly stretched value to a value below S_L . Contrary to expectation, the CH₄/air flame kernel with LFV of 41 cm/s grew faster than the C₂H₄/air kernel with LFV of 51 cm/s. From the normalized propagation speed S_{y*}/S_L , it was observed CH₄/air experienced the most stretch at development stage and least stretch at the late kernel stage.
6. At lower flame temperature (i.e., 2134 K AFT), the observed fuel impact on Y_* increased which agrees with the mixture Lewis Numbers. This observation also agrees with previous research conclusion that mixtures with higher Lewis Number would require a larger critical radius to development into a sustained flame kernel [49].
7. It was also observed that at lower flame temperature (i.e., 2134 K AFT), S_{y*}/S_L shows that the sensitivity to stretch widened between CH₄/air and the rest mixtures due to wider variation in both the Markstein length and effective Lewis Numbers.

Effects of variation in Fuels with constant LFV

1. For selected mixtures with LFV of 41 cm/s (i.e., methane/air, propane/air, and ethylene/air), propane kernel developed at slightly lower rate than methane and ethylene kernels which grew at the same rate.
2. Analysis of the image peak OH* intensities at LFV of 41 cm/s showed that ethylene was the most reactive, while methane and propane was about the same. Dilution of the flows (i.e., at LFV of 29 cm/s), resulted in an overall drop in intensity, however the decay was more significant in propane and ethylene. The higher loss in propane and ethylene may be due to an increased stretch on

the flame temperature away from the AFT which results from wider variation of Le of the fuel in the lean range [162].

3. Analysis of the integrated OH^* intensity at LFV of 41 cm/s showed ethylene/air mixture to have the strongest kernel while propane/air mixture had the weakest kernel. As earlier, the decay in diluted mixtures with LFV of 29 cm/s was more significant in propane and ethylene.
4. Analysis of the axial tip locations of the flame fronts showed a symmetric growth curve at all mixture conditions investigated with propane kernel having the least growth. At 41 cm/s, the propagation of methane and ethylene was nearly the same, however the methane kernel propagated faster under leaner mixtures at the LFV of 29 cm/s. This change in characteristics may be due to the increased differences in their Lewis Numbers.
5. The general trend in S_{y+} shows that the propagation rate of the flame front decreases asymptotically from a highly stretched value to a value below SL with propane kernel having the lowest value. From the normalized propagation speed (S_{y+}/S_L) of mixtures at LFV of 41 cm/s, the kernel in CH_4 /air was more stretched than that of C_2H_4 /air at the development stage. At a lower LFV of 29 cm/s, the CH_4 /air kernel became more stretched than the C_2H_4 /air kernel. This change in trend may have resulted from higher stretching of the C_2H_4 /air flame temperature from the AFT which is also depends on the Lewis number of the deficient component (i.e., the Fuel).

Chapter 7 Discussion of Overall Results

7.1 Introduction

As stated in the introductory chapter, this PhD work involves detailed investigation of the development characteristics of a LISI flame kernel in premixed fuel/air mixtures. The goal is to show the key development characteristics of the LISI flame kernel and possibly link them to ignition success or failure. It is also anticipated that the data from this investigation could be used for development and validation of laser ignition models. This goal is accomplished through several experimental campaigns the results of which were used for both visual characterisation and parametric study. The parametric study also involves dynamic analysis of the kernel geometric growth, the flow velocity field, and the intensity of the emitted OH* all of which have been reported in chapter 4, 5 and 6. The purpose of this chapter is to collectively discuss all the results from the investigation in a coherent and insightful manner.

7.2 Visual characteristics of the Flame Kernel development

The development of the LISI flame kernel from the ignition point to 1667 μs time delay is captured by tomographic images shown in Figure 4.1a - Figure 4.1b. Further visualisation of the kernel development is also obtained for 50 μs to 10 ms time delays using the mean OH* chemiluminescence images shown in Figure 5.1 - Figure 5.2. Both visualisations reveal that the LISI flame kernel starts as an elliptically shaped plasma of hot gas which then evolves into a two-lobe toroidal flame front, followed by the appearance of a front lobe leading to toroidal asymmetry and finally a self-sustaining flame ball. They also provided a clearer picture of both the fluid dynamics and reactivity of a developing LISI flame kernel.

Dynamics of the Kernel geometry

At the start of ignition, the first feature observed from the tomographic images is an elliptically shaped plasma of hot gas formed within the mixture. This hot plasma usually between 1-3 μs and is accompanied with shock waves. Many authors [92],[20][100] observed that the plasma usually extends more towards the direction of laser beam and the shockwave centroid is usually displaced from that of the plasma.

The second feature observed at 50 μs from both image sets is a two-lobe toroidal flame kernel propagating symmetrically above and below the laser axis. Previous studies observed similar shape for both laser ignition and electric spark ignition [92],[100]. The origin of this feature has been attributed to an induced inward flow resulting from an over-expansion of the shock wave, emanating outward from the region of the discharge at a very high velocity [138]. Figure 2.8a - Figure 2.8b provides an illustration of the transformation from ellipsoidal spark kernel to a toroidal flame kernel from 1 to 50 μs . As shown by the figure, two contra-rotating vortex rings (indicated by velocity streamlines of magnitude 25 m/s) are generated in the upstream and downstream regions via a complex process of interaction of the shock waves and rarefaction waves from the hot plasma. The inward flow from these vortices leads to the formation of the toroidal flame front. The figure also suggests that the leading-edge vortices (directed towards the laser) is smaller in size which implies it decays more rapidly than the trailing edge vortices.

The third feature observed at 100 μs is the appearance of a front lobe which propagates in the direction of the laser, thus leading to an asymmetric toroidal flame front. Previous studies [92] has suggested that this is a unique feature of laser ignition. Although observed by several authors, it is not yet clear how this feature develops. For example, Spiglanin *et al.*, 1995 [92] suggested that it might be due to the initial flow field created by the propagation of a radiation transport wave up the laser beam, arising from the high rate of energy transfer at the leading edge of the plasma. While it is true that the plasma kernel created by laser breakdown can result in an ionized front propagating toward the laser [155],[20] this is not expected to continue long after the laser pulse had ceased, hence, there

is no evidence of this. Following from the differences in decay rate between the leading edge and trailing edge vortices, Bradley *et al.*, 2004 [20] suggested this phenomenon is capable of producing a centreline velocity towards the laser source, leading to generation of the front lobe. Figure 4.3 Figure 2.10 clearly illustrates this transformation in the superposed kernel edges formed between 50 and 166 μs . As shown in the figure, after the weak leading-edge toroid has dissipated, the induced gas velocity to the left, up the laser beam, can be high. This results in a flow of hot gas close to the centreline to the left and its displacement by cold gas from the right. The elongated hot gas kernel comprises the third lobe. This displacement of the centreline hot gases by the cold gases is clearly shown in the observation images following the appearance of the front lobe.

The final shape of the propagating flame front is that of a typical flame ball following local quenching at the front lobe. This is shown in Figure 5.2 which represents the sequence of mean OH^* chemiluminescence images acquired between 3 and 10 ms time delays. As shown by the figure, propagation of the flame front is marked by changing radius of curvature in both the upstream and downstream directions.

Dynamics of the Kernel reactivity and reaction zones distribution

An investigation of the colour scales of the OH^* chemiluminescence images (Figure 5.1 - Figure 5.2) reveals that the reaction intensity changes progressively during transition to a self-sustained flame front. Between 50 μs and 100 μs , the peak intensity of OH^* dropped from 3800 AU to 2300 AU due to cooling of the hot gases which make up the initial kernel. At 150 μs , a new peak in OH^* intensity occurs which is the first indication of the kernel survival. Subsequent kernels showed a continuous decay in peak OH^* until a minimum peak is attained beyond which the peak rises again following the formation of a self-sustaining flame. These would be shown further by quantitative analysis of the OH^* intensity.

In addition, the image colourmaps also reveal that the distribution of reaction zone changes at different stages of development. At the initial stages (50 - 300 μs), the OH^* is evenly distributed, hence the kernel is composed of an isotropic reaction zone. A possible explanation for this is that initial kernel growth is predominantly

due to the presence of hot gases which originated from the initial hot plasma. At the mid stage of development (450 - 1500 μs), the distribution of OH* reveals the kernel is composed of anisotropic reaction zones with peak intensity within the middle region. A possible explanation for this is that kernel growth during this period is the result of both induced gas flows and chemical reactions. This could also be the result of mixing of the reaction zone with the flow due to vortex induced motion which may lead to multiple flame fronts within the kernel. As shown in Figure 5.2, the colourmap of the late kernels (3 - 10 ms) shows that the reaction zones (i.e., flame fronts) were concentrated at the edges. Unlike the early kernels, the inner region of low intensity may indicate the presence of burnt gases. These observations also give an idea of the two-way interaction involving the flame-flow mixture and flame-burnt products.

7.3 Analysis of the Flame Kernel growth and propagation

The first output from geometric measurements following edge extraction of the tomographic images is the initial plasma radius. The result reveals that following generation of the spark at ignition, there is nearly an instantaneous growth of the plasma to an initial finite size. This growth has been attributed to the sudden energy deposition at breakdown which generates high pressure in the kernel leading to a rapidly expanding wave[90]. Therefore, the growth of the plasma at this stage can be predicted using the spherical blast wave theory [91], with the assumption that heat release due to chemical reactions is negligible. Table 4.2, compares the measured plasma radii and prediction at 1 μs based on spherical blast wave model (equation 2.15), assuming 70% spark energy deposition efficiency. From this analysis, it was shown that the predicted plasma radius was 13.9% more than the measurement laminar case and 4% less than measurement in turbulent case. The observed differences in the initial sizes of the kernel for both conditions despite having similar pulse energy highlights the fact that laser induced gas breakdown is a stochastic process, hence similar pulse energies can produce different spark energies, although the same spark efficiency of 70% has been assumed in the prediction.

The second output from the measurements is the displacement of the flame front tip along opposite directions of the flow axis. Figure 5.6 shows that the downstream flame front of stoichiometric CH₄/Air is characterised by non-linear decay growth during the first 0.8 ms, followed by a steady linear growth onwards. Similarly, the upstream flame front follows the same trend until a maximum displacement of 12 mm at 7 ms is reached. The combined history of both measurement (i.e., Y₊ and Y₋) reveals a nearly symmetric growth curve about the laser axis, however this did not appear to be the case in the 2D scatter shown in Figure 5.5. A comparison of the axial growth curve at different flow velocities (i.e., 1.15, 2.08, 3.0 and 3.48 m/s) is shown in Figure 5.20, which reveals that the loss of symmetry is due to displacement of the kernel centroid by the flow which means symmetric growth should be obtained if ignited in quiescent mixture flows. When fuel lean mixtures (i.e., $\phi = 0.9, 0.8$) are compared with the stoichiometric mixture, the kernel growth is slower in lean mixtures and the maximum displacement reached by the flame front upstream is lower as shown in Figure 6.7. This is expected considering the differences in laminar flame speed of the mixtures. It also shows a higher strain effect of the flow on the flame front of leaner mixtures. An interesting result is the comparison between three fuels (methane, propane, and ethylene) with the same AFT ($T_{ad}=2226$ K) in which propane experienced the slowest growth while methane was the fastest. This is unexpected because ethylene has the highest estimated laminar flame speed (i.e., $S_L = 3.87$ m/s for C₂H₄/air) while methane has the least value (i.e., $S_L = 3.06$ for CH₄/air). It is possible the observed differences may be the result of preferential diffusion effects which occurs in mixtures with non-unity Lewis number values. As shown by the results, propane with the highest Lewis number ($\phi=0.94, Le=1.4$) is the experiences the slowest growth while methane with unity Lewis number (i.e., $\phi=1.0, Le=1$) experiences the fastest growth. This is in agreement with observation by past authors [161], that mixtures with higher Le values would require a larger critical radius for the flame to be sustained. In other words, the CH₄/air mixture with the least Le value would require lower **MIE** and less time to develop into a self-sustained flame kernel compared with the C₃H₈/air mixture. Hence, the order of duration in reaching a self-sustainable flame kernel is methane, ethylene, propane. A similar trend in axial flame tip displacement is observed in leaner fuel/air mixtures

(CH₄/air @ $\phi=0.9$, C₃H₈/air @ $\phi=0.86$ and C₂H₄/air @ $\phi=0.78$) with $T_{ad}=2134$ K as shown by Figure 6.22. However, the differences become wider which is in accordance with their Le values. For fuels heavier than air (e.g., C₃H₈, C₂H₄) Le values increases with reduced ϕ values while the reverse is the case for fuels lighter than air (e.g., H₂, CH₄)[162]. Further analysis revealed that at lower AFT ($T_{ad}=2134$ K), the differences in displacement between the three mixtures widened which is in accordance with their Le profiles the fuels.

Unlike the two axial flame fronts, Figure 5.6 shows that the displacement of flame front towards the laser is characterised by sudden change in slope at 1.5 ms due to local quenching at the front lobe, followed by a steady linear growth onwards. As a result, measurements of the kernel dimensions reveal that the height of the flame kernel (which was measured along the flow axis) is always greater than the width (which was measured along the laser axis). This can be seen in Figure 5.7 which further reveals there is small differences between them in the first 1.5 ms due to the presence of the front lobe and the asymmetry in the initial spark dimensions [97]. Beyond 1.5 ms, the difference increases due to variation how the flow velocity affects growth along different directions. Based on the orientation of the laser and the flow condition investigated, the kernel expanded to reach the size of the burner (i.e., diameter =22 mm) in 2.5 ms, whereas it would have taken about 1.5 ms to cover the same space if it were oriented transversely. A comparison of the transverse dimensions of the kernel (W) at different pulse energies is shown in Figure 5.14 which reveals an enhanced early growth due to higher energy, however no significant changes were observed in the fully developed kernels. It is worth noting how W increased significantly during the first 1 ms when the incident energy increased from 32 mJ to 64 mJ, however, further increase in energy had little or no effect in the size. This shows the existence of a threshold incident energy beyond which increase in pulse energy had no impact on the growth. The observation agrees with previous authors that spark energy deposition has a limiting efficiency depending on the lens focal length [90],[157]. At 10 ms time delay, no significant changes were observed in the kernel growth irrespective of the magnitude of the incident pulse energy. Further details of the sensitivity analysis can be found in Table 0.1 (see Appendix E: Sensitivity Analysis).

Another common characteristic evidenced in all the cases studied is that the propagation rate of the flame fronts is marked by decay from a highly stretched initial value to a steady asymptotic value close to the unstretched laminar flame speed. Figure 5.8 reveals that as the kernels develops, the propagation rate of the downstream flame front decreases until it converges to a value slightly above the unstretched laminar value, S_L . In contrast, the propagation rate of the upstream flame front continues to decrease after reaching the unstretched value S_L before converging to its asymptotic values. The difference in growth rate between the two flame fronts implies different levels of stretch which could be due to several factors. One of the factors could be differences in the surface area of the flame fronts as a result of wrinkling induced by flow turbulence [163]. Another factor could be that the local S_L values have changed due to varying compression of the mixture in both the upstream and downstream region by the expanding flame kernel [90]. This figure also highlights the existence of three key phases of steady growth. For example, the first steady growth of the downstream flame front was observed between 150 to 200 μs , when the flame front propagation rate was 11 m/s following the appearance of the front lobe. This growth dominated by induced gas velocity of the shock waves. The second steady growth occurred between 250 and 500 μs , when the kernel propagation rate was 7.5 m/s which marked the formation of an igniting kernel. This growth is due to combined influence of chemical reactions and induced gas velocity. The third steady growth occurred from 800 μs onwards when the flame front propagated at its asymptotic velocity of 3.5 m/s. At this stage, the growth is purely by chemical reactions when variation from its laminar unstretched value (i.e., 3.06 m/s) is at its minimum. A comparison of the growth rate at different incident energies (Figure 5.15) reveals that increase in energy of the incident beam only enhanced the early kernel growth before 1 ms. A clearer picture of stretch effects due to increase in pulse energy is shown in a plot of the normalised value versus the downstream displacement (Figure 5.16). This reveals that for a displacement of 8 mm, the respective stretched speed at 32, 62 and 128 mJ were 20, 20, 30, and 50% more than the upstretched laminar value. The similar stretch between the two flame fronts at 96 and 128 mJ further confirms the existence of a threshold incident energy above which further increase has no impact on the growth. As shown in Figure 6.11 - Figure 6.12 the axial

propagation rate of the flame fronts is slower in lean mixtures (i.e., $\varphi = 0.9, 0.8$) compared with the stoichiometric mixture. Analysis reveals that the effect on dilution on the steady state S_{y+} values is a linear decline in laminar flow situation (i.e., 11% drop per unit reduction in φ between $\varphi=1.0$ to $\varphi=0.9$), however, the variation is non-linear in turbulent mixtures. It was also observed that the stretched flame speed appears to be slightly higher under lean conditions. Further analysis of the growth rate normalised by the unstretched laminar flame speed (Figure 6.13 - Figure 6.14) showed higher stretch rates in leaner mixtures compared with the stoichiometric. The mixture with the least φ value experienced the most stretch. This may be due to the fact that the Le values of lean CH_4/air mixtures are slightly below unity, therefore preferential diffusion may affect growth. The clearest evidence of the effect of preferential diffusion on growth rate is observed when is compared for three different fuels at constant AFT (Figure 6.25 - Figure 6.26). The results clearly shows that fuels with higher Le values were marked with higher variation from the unstretched laminar flame speed.

7.4 Analysis of the OH^* Intensity Characteristics

Analysis of the spatially integrated OH^* intensity provided information about the heat release strength of the flame kernel as well as further evidence of an important transition point in the lifecycle of the kernel. Figure 5.10 shows the integrated OH^* intensity history of the kernel for stoichiometric CH_4/air in laminar flow condition. An important feature of this graph which appears in all cases studied is that it involves an initial decline to a minimum trough value. This occurred within 200 μs in the stoichiometric CH_4/air mixture with a trough value of 44000000 AU. This initial trough could be interpreted as the transition point between a quenching flame kernel and the formation of an igniting kernel. The time elapsed before this transition can be interpreted as the chemical induction period [92]. The results show that the duration changes systematically depending on the incident energy, equivalence ratio and the fuel mixture. For example, the induction time increased with higher incident energy leading to lower trough values. For different equivalence ratio (i.e., $\varphi = 1.0, 0.9, 0.8$), the duration also increased in leaner mixtures but did not lead to lower trough values. Of the three

fuels investigated, the shortest duration occurred in methane despite ethylene having higher values.

As the kernel continues to develop beyond this transition point, the cumulative heat release rises which is marked by a continuous rise in the integrated OH* intensity. A rising trend in the integrated OH* intensity also shows that the developing kernel has a higher chance of survival. In all the cases studied, the rising trend reaches a peak and is followed by a downward trend which may be due to heat losses resulting from interaction of the flame kernel with the Nitrogen used as blow-off gas. In the stoichiometric CH₄/air mixture, this peak appeared in the form of a plateau with intensity value of 6000000 AU which occurred between 6 ms and 8 ms. Sensitivity analysis shows that the peak integrated OH* intensity differs systematically depending on the incident energy, equivalence ratio and the fuel mixture. For example, the maximum integrated OH* intensity decreased when the incident energy from 32 to 64 mJ, however, further increase in energy produced an increase but not at the same as the initial peak with 32 mJ. Based on this observation, the optimal ignition energy should be decided based on the OH* production. In the present investigation, a 32 mJ energy would be ideal, however, it will be helpful to investigate lower incidents energies since this is much higher than the MIE of the mixture (~0.25 mJ). For different equivalence ratio (i.e., $\phi = 1.0, 0.9, 0.8$), the maximum integrated OH* intensity decreased in leaner mixtures. Of the three fuels investigated, the maximum integrated OH* intensity was obtained in ethylene, followed by methane and then propane. This shows that a higher heat release was occurring in ethylene kernel despite the faster growth from the methane kernel.

Further analysis of the peak OH* intensity history revealed that transition to a self-sustained flame kernel is characterized by decay from an initial high peak OH* intensity to a minimum value which must be sustained for the kernel to survive. This is shown in Figure 5.11 and is also evident in all the cases investigated. According to the figure, after reaching its minimum value at 1.5 ms, a slow recovery stage is observed with subsequent peak OH* intensity reaching a maximum trough value of 1500 AU at 4.5 ms and a second maximum trough value of 2000 AU at 9 ms. The high decay rate in peak OH* intensity during the early

development of the kernel is indicative of the cooling resulting from high stretch on the kernel. The recovery indicates continuous heat release resulting from chemical reaction at the flame fronts. Just like most of the parameters, sensitivity analysis showed that the minimum peak OH* intensity changed systematically depending on the incident energy, equivalence ratio and the fuel mixture. As observed in the integrated OH* intensity, the values of the peak OH* intensity decreased with energy increase from 32 to 64 mJ following the formation of a self-sustaining flame. However, further increase in energy however did not produce any further change as observed in the kernel width. As expected, the magnitude of peak OH* intensity decreased in leaner mixtures (i.e., $\phi = 0.9, 0.8$) compared to the stoichiometric mixture (i.e., $\phi = 1.0$). Of the three fuels investigated, the values of peak OH* intensity shows ethylene was the most reactive while propane was the least reactive. However, the reactivity of methane kernel approached that of ethylene when the mixtures were leaned further.

Chapter 8 Conclusions and Recommendations

8.1 Conclusions

This thesis report investigated key development characteristics of laser induced spark-ignited flame kernels propagating in gaseous premixed fuel/air mixtures. The goal was to show the link between the characteristic parameters and ignition success with the hope that the results could be used as validation data for laser ignition models. Two main studies of the kernel development were explored through visualisation of flow field features (Laser tomography) and flame chemiluminescence. The third investigation was done mainly to show the sensitivity of the parameters to changes in the fuel properties. As a result, two experimental measurement solutions were developed. The first setup consists of a co-axial straight tube burner in which flowing mixture of Methane and Mie-scattered Air was illuminated by a laser-sheet and then ignited by a single pulse laser beam of 1064 nm wavelength and 7 ns pulse width. High-speed imaging of the combustion region allowed simultaneous measurement of the flame kernel development as well as its flow field structures. The second setup consists of an axial contracting-nozzles burner in which different flowing fuel/air mixtures were repeatedly ignited using a focussed laser beam of 1064 nm wavelength and 2 Hz frequency. Direct imaging of the flame kernel OH* chemiluminescence using intensified CCD camera. Conclusions from the findings are summarised below:

- High speed flow visualization study revealed that on ignition, an elliptically shaped plasma of hot gas was formed, which developed into a two-lobe toroidal kernel by 50 μ s, followed by the appearance a front lobe before 167 μ s, thus resulting in an asymmetric toroidal flame kernel. The images also reveal an induced motion of a centreline jet flow, generated by contra-rotating upstream and downstream vortices which was responsible for the front lobe appearance. Analysis of the initial plasma reveal an instantaneous growth, the radius of which is comparable to that predicted by the Blast wave theory at 1 μ s with

some differences depending on the flow (i.e., 13.9% less in the laminar flow case, and 4% more in the turbulent flow case). Analysis of the spatial growth revealed that in the laminar flow case, it took about 0.7 ms for the kernel height to grow to a size about the burner diameter (i.e., 10 mm) and while in the turbulent flow, it took only 0.3 ms to reach this height. Further analysis of the growth rate revealed it decayed in stages from an initial shock speed 23 times the unstretched laminar value (S_L) to a stable speed close to S_L in about 1 ms.

- Prior to ignition, PIV analysis reveal an axisymmetric structure in the axial flow velocity vectors, with peaks in the potential core region which decreased radially to its minimum values near the flow edges. From the radial profiles, a constant velocity peak was observed throughout the flow in the laminar flow case, while in the turbulent flow the velocity peak decreased moving from upstream to downstream. Unlike the flow velocity, the turbulence intensity was stronger near the flow edges than in the potential core region with an increasing trend in magnitude from upstream to downstream. Analysis of the flow field after ignition show an increased peak velocity in the downstream flow ahead of the flame front and reduced peak velocity in the upstream flow behind the flame front. Further investigation of the radial profiles of the axial velocity revealed that this variation in velocity peak existed only within certain distances (below 2.5 mm) from the flame fronts.
- Flame kernel visualisation using the OH^* chemiluminescence revealed transition from a toroidal shaped kernel at 50 μs , to a toroid with front lobe at 100 μs and lastly a self-sustained propagating flame ball after disappearance of the front lobe. The displacement curve of the flame fronts reveals two stages in growth: a non-linear decay growth in the first 0.8 ms, and a steady (linear) growth onwards. The dimensions of the kernel were different with the height always greater than the width. In the base case, it takes about 2 ms for the kernel height (H) to grow to a spatial scale about the size of the burner diameter (i.e., 22 mm) and kernel width (W) reached a spatial scale about the size of the burner diameter in just above 2 ms. The propagation rate of the flame front decays from a highly stretched initial value to a steady asymptotic value close to the unstretched laminar flame speed. It also highlights three key

stages in development of the kernel. In the base case (i.e., laminar CH₄/Air at $\phi=1.0$), the first steady velocity of 11 m/s dominated by induced gas flow was observed between 150 to 200 μs . The second steady velocity of 7.5 m/s was observed between 250 and 500 μs due to combined influence of chemical reactions and induced gas flows. The third steady velocity of 3.5 m/s, observed from 800 μs and beyond was purely due to chemical reactions.

- Further visualisation of the image colourmap revealed the spatial distribution of OH* produced inside the reaction zone evolved from isotropic to anisotropic flame front during its development. In the self-sustained flame, the reaction zones were concentrated at the edges. Transition to a self-sustained flame kernel is characterized by decay in peak OH* intensity to a minimum value which must be sustained for the kernel to survive. The kernel development involves transition from a quenching kernel of hot gas to the formation of an igniting flame kernel which is marked by a minimum integrated OH* intensity. The time elapsed before this minimum value was interpreted as the chemical induction time [92]. In the base case, this transition occurred at 0.2 ms. Following this transition, the kernels with a high chance of survival is characterised by a continuous rise in integrated OH* intensity attaining a maximum value when the kernel becomes fully developed.
- A comparison of the kernel development at different incident energies (i.e., 32, 64, 96 and 128 mJ) reveals that the early growth of the kernel is enhanced by a higher energy, but this happens within a limiting threshold energy. In the fully developed kernel however, the size of the kernel is not influence by the energy. A comparison of the peak OH* intensities reveals that the reactivity was unaffected by the energy of the incident beam except in the first 0.5 ms. A comparison of the integrated OH* intensities shows that the chemical induction duration increased for higher energies. An interesting insight from this analysis is the fact that the maximum integrated OH* intensity decreased greatly by 25% with energy increase from 32 to 64 mJ, however, further increase in energy resulted a reduction in the negative effect. Therefore, in choosing the energy, the OH* production is very important since stretch effect only last for a short time. For the current investigation, 32 mJ would be the best choice, on the

observations, should be decided based on, however further investigation with lower ignition energies is advised in order to decide the optimal ignition energy. Further details of the sensitivity analysis are shown in Table 0.1 of Appendix E: Sensitivity Analysis.

- A comparison of the kernel development at different bulk flow velocities (i.e., 1.15, 2.08, 3.0 and 3.48 m/s) reveals an increased displacement of the kernel centroid with higher flow velocity due to convective effects and as well as an increased strain on the flame front upstream. Overall, there is no remarkable increase in growth rate of the kernel due to increased velocity. The combined effect of convection and increased turbulence at higher bulk velocities resulted in a faster propagation of the flame front downstream. There were fluctuations in both the peak OH* intensity and maximum integrated intensity which made difficult to understand the influence of increased velocity on reactivity. Also, the duration of chemical induction appears to be longer for higher bulk velocity flows.
- A sensitivity study of the effect of equivalence ratio on kernel development reveals a linear decline in the characteristic parameters moving from $\phi=1.0$ to $\phi=0.8$ with higher stretch rates in leaner mixtures. For example, the steady state values of S_{y+} in the laminar mixtures were 3.3, 2.96 and 2.59 m/s, which represents 11% drop per unit reduction in ϕ , moving from stoichiometric to lean. Also, the minimum peak OH* intensities were lowered by 18% per unit reduction in ϕ during its development. In addition, the integrated OH* intensities showed that lower equivalence ratios produced weaker kernels (9% per unit reduction in ϕ) and experienced longer chemical induction duration. It is worth noting that the linear relationship did not occur in turbulent flows.
- A sensitivity study of the variation in kernel development characteristics for fuels with the same AFT showed significant differences depending on the mixture Lewis number, Le . Of the three fuels investigated, the kernel experienced the slowest growth in propane fuel with the higher Le . Contrary to expectation, kernel growth was faster in methane (with LFV of 41 cm/s) than in ethylene (with LFV of 51 cm/s) due to preferential diffusion that occurs in mixture with non-unity Le . From the normalized propagation speed S_y-/S_L , it

was observed CH₄/air experienced the most stretch. The differences in stretch widened with lowering of the AFT from 2226 K to 2134 K due to wider variation in both the Markstein length and effective Lewis Numbers. Both peak OH* intensity and the spatial integrated OH* intensity measurements showed the ethylene kernel was the most reactive while propane was the least reactive. Likewise, the duration of chemical induction followed the same order with ethylene kernel taking longer than the rest.

- Sensitivity study using fuels with constant LFV showed some difference both geometric stretch and reactivity. At 41 cm/s LFV, the ethylene kernel was the most stretched while the propane kernel was the least stretched. However, further dilution resulted in methane becoming more stretched. The same trend is observed in reactivity as shown by both the peak OH* intensities and integrated OH* intensities. This behaviour may be explained by the fact that there is more heat losses as the flame temperature difference is expected to widen in line with wider variation of Le of the fuels in the lean range [162].

8.2 Recommendations and Further Work

- Laser tomography was employed for simultaneous visualization of the flame kernel and its flow field. From this the flame kernel characteristics such as flame tip positions and displacement speed were quantified. To effectively describe the flame kernel, a complete mapping of the characteristics of the entire kernel geometry at different times is necessary. In addition, repeated imaging of the flame kernel at the same condition would be helpful in statistically defining the reproducibility.
- High Speed PIV was employed to obtain the flow field structure before and during ignition. In the isothermal case, the velocity fields were based on an average velocity, however only instantaneous velocity field were obtained during reaction. To statistically define the reproducibility of change in velocity field during ignition, it is necessary to acquire more high-speed images at the same condition and then find the mean using phase averaging technique. In

addition, a more realistic picture of the flow structures could be obtained by multi-camera PIV technique.

- OH* chemiluminescence was employed as flame marker to characterise the flame kernel as well as to indicate the relative measure of heat release. To effectively characterise the flame kernel, it would be helpful to measure the chemiluminescence of other radical (e.g., CH*, CO₂) produced in the reaction. In addition, a better picture of the 2D geometry of the flame kernel could be obtained from more sophisticated techniques like PLIF.
- Differences in the flame kernel characteristics due to changing pulse energy and flow velocity were observed in this work. Characterising the sparks produced by the laser beam under these conditions would be helpful in explaining the reason for these differences since the actual energy utilized in laser ignition comes from the spark.
- Differences in the flame kernel characteristics for different fuels were observed in this work. It would be interesting to look at the actual effect of the chemical components such as the carbon size and bond type. In addition, studies on a wider range of low emission fuels would be helpful in developing more robust laser ignition models and the work towards engine-relevant conditions.
- This research was conducted with the hope that it will produce useful data for improved modelling of laser ignition process. Further research in this area would be more useful if there is close collaboration with modellers such that specific data required to validate existing codes could be produced.

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Appendices

Appendix A: Experimental Procedures

{Note: These procedures were developed for a broader research work including spark ignition work not reported in this thesis}

Prior to conducting the experiments:

- A leakage test is done by opening the gas supply line (air, methane, hydrogen or carbon dioxide) and applying a leak identifying agent to potential leaking points like connections between pipes.
- A visual scan is then performed to make sure there are no flammable objects near the ignition location.
- The Extraction system is positioned above the burner to suck any unburnt fuel out of the laboratory.
- A confirmatory test should be performed to ensure the electrode gap can produce consistent sparks in open air (electric ignition).
- Ensure the laser beam is optically aligned and a confirmatory test should be performed to ensure consistent sparks in open air (laser ignition).

The experimental procedure is based on the flowchart presented in Figure. Based on this flowchart, the sequence of activities to be performed are described below:

Start up

1. Ensure the Ignition unit (or Laser system), control and measurement Instruments are switch ON with the correct settings.
2. Open the gas cylinder regulator valves and set delivery pressures to 1 bar.

Gas flow setup

3. Open the Nitrogen valve to purge the system of combustion products and unburnt gases accumulated from the last experiment. Turn off the Nitrogen.
4. Open the Air Flow Meter to supply the desired amount of Air. Record the air flow rate.
5. With the Solenoid Valve (SV-1) ON, open fuel valves to set the correct fuel flow rates for the desired mixture equivalence ratio and exit flow velocity.

Safety check

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6. With the flow rates correctly set, quickly turn OFF the Solenoid valve (SV-1 in Figure 1) to stop the fuel flow.
7. If using electric ignition source, ensure high voltage warning sign is placed at the door.
8. If using laser ignition source, ensure the door safety interlock switch is activated and a laser warning sign is placed at the door.

Gas mixing, Ignition and measurement

9. With the air flow ON, switch ON the Solenoid valve to initiate the fuel flow.
10. Wait for 15 seconds for the air and fuel to mix.
11. Ignite the mixture and initiate measurements by pressing the Spark Trigger button.

Spark Energy: the spark energy is obtained from the oscilloscope display of the transient Voltage and Current triggered by the spark signal. The pulse energy output from the laser is recorded by using an Energy meter.

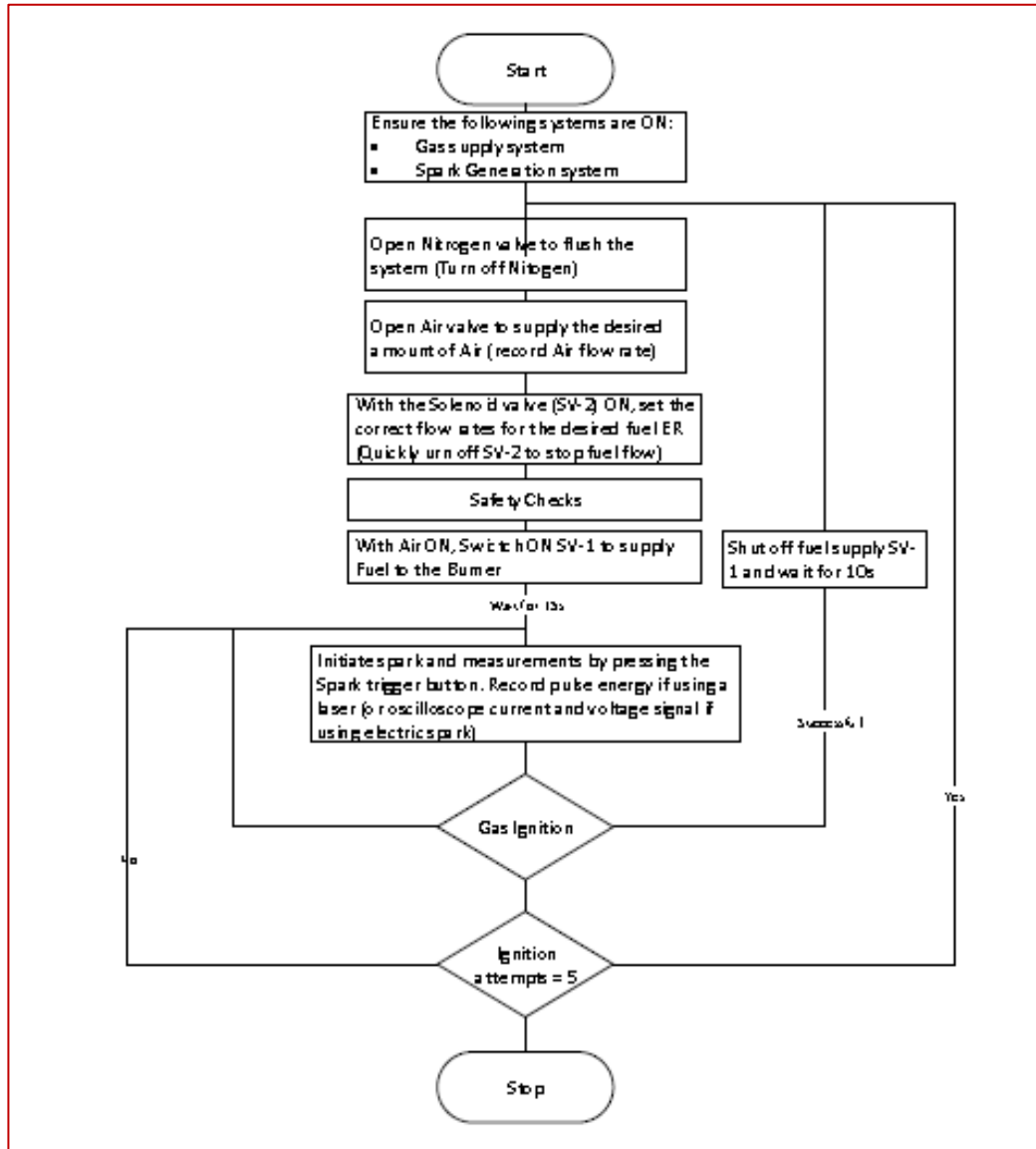
Image acquisition: The development of the flame kernel is recorded by the TSI camera.

12. Switch OFF the Solenoid valve to stop the fuel flow.
13. If the gas mixture is ignited by spark (as observed through visual observation or chemiluminescence) then, purge system with Nitrogen (step 3) and repeat experiments with fresh gas air mixture (steps 16, 17, 18) for specified number of runs (say 20 runs)
14. If the gas mixture did not ignite, then increase the spark energy either by increasing the pulse width (electric ignition) or reducing the Q-Switch delay (laser ignition)
15. Repeat process for different equivalent ratios, flow velocity and mixture composition.

16. Shutdown

17. Switch OFF the Ignition unit (or Laser system), control and measurement Instruments.
18. Close the gas cylinder regulator valves and de-pressurize lines.

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Appendix B: Risk Assessment

{Note: This Risk Assessment was developed for a broader research work including spark ignition work not reported in this thesis}



Activities, Hazards, Controls

Reference: RA015952/1

1. Laser ignition experiment in Thermodynamics Laboratory

Sign-off Stat

<p>Description of Activity:</p>	<p>This is an experiment where different Hydrocarbon-Air mixtures are ignited in a Co-flow Burner using both a laser and electrical ignition systems. The laser system comprises a Q-switched Nd-YAG laser (Class 4) focused on the Burner exit via a collection of optics in space. The electrical spark is generated between two electrodes (at the burner exit) from the high voltage side of an ignition coil which is powered by a 12 volts car battery.</p>	
<p>Hazard 1. Laser beam</p>		
<p>Risks:</p> <ul style="list-style-type: none"> - Eye injury/damage, Skin burns, etc from contact with lasers and diffuse reflections - Ignition of flammable materials - heating/vaporization of materials causing hazardous fumes <p>Risk level: - Medium</p>	<p>Existing Control Measures</p> <ul style="list-style-type: none"> - Beam dump to terminate beam at the end of its useful path - Black Shield cover to conceal and absorb the area from stray radiations - Door interlock system in place - The laser equipment is supported on a fixed platform so as avoid toppling - Beam paths kept below eye level. - Adherence to instructions on the Operational Manual - Eye protection to be worn during operation - Notices and sign placed outside the door 	

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Hazard 2. Compressed Gases (e.g. Methane, Hydrogen, Nitrogen and Carbon dioxide)	
<p>Risks: -Methane: Methane is a colorless and odorless gas. It forms flammable and explosive mixture with air between 5%-15% concentration. Asphyxia may result if the oxygen concentration is reduced below 18% by displacement.</p> <p>- Hydrogen: Hydrogen is a colorless and odorless gas. It forms flammable and explosive mixture over a wide range of concentration (5%-75%). It burns with almost an invisible blue flame. Primary health hazard is Asphyxiation by displacement of Oxygen. - Over-exposure to inhaled aerosol Risk level: -Medium</p>	<p>Existing Control Measures</p> <p>- Fumes/Gas detection and extraction systems are installed - Solenoid valves, flashback arresters are installed on the fuel delivery line - Gases are stored in isolated and well ventilated cupboards - Only small quantities of flammable gases will be used for experiment - The fuel lines are operated at atmospheric pressure such that only minor leaks is envisaged in case of valve malfunction Leakage test is done at potential leakage points - Visual scan to ensure no flammable objects nearby - safety valves, flow regulators and flash back arresters are in place</p>
Hazard 3. Hot surfaces (e.g Gas Burner, Electrodes)	
<p>Risks: - Burns from contact with hot surfaces - Explosion through contact with flammable gases Risk level: -Medium</p>	<p>Existing Control Measures</p> <p>- System is ignited and turned off in a controlled manner - Hot surfaces/materials allowed to cool before handling</p>

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Hazard 4. Electrical Hazards (e.g. Power supply units including high voltage power to Laser)	
<p>Risks: - electrical shock from electrical equipment and exposed cables - death from electrocution - fire and burns Risk Level: - Low</p>	<p>Existing Control Measures</p> <p>Both the Laser and Electric ignition systems are operated in a controlled manner The electrical cables are carefully checked for signs of wear/tear, etc. The earth connection is connected for all the electrical equipment. Appropriate fuses are placed in each piece of equipment The room is an explosion proof room. Facilities controlled by UCL staff</p>
Hazard 5. Environment	
<p>Risk: Risk level -Slip, trips and falls : medium risk - Inhalation of poisonous fumes or particulate matter: medium risk -Hearing damage due</p>	<p>Existing Control Measures</p> <p>- Proper house keeping - Extraction system and ventilation system in place - PPE (Ear plugs)</p>
Hazard 6. Spark Radiations	
<p>Risks - Retina damage due to intense spark light - cataracts, cornea damage due to infrared radiation - skin burn, blurred vision due to UV radiations Risk level: Medium</p>	<p>Existing Control Measures</p> <p>Wearing proper eye protection</p>
Hazard 7. Manual handling	
<p>Risk: back injuries, shoulder pain due to occasional movement of burner, imaging and optical equipment Risk level: low</p>	<p>Existing Control Measures</p> <p>proper lifting; not working in one position for long periods of time</p>
Hazard 8. Lone/out-of-hours working	
<p>Risk: -delay in getting help in the event of emergency, - being attacked by intruders. Risk level: low</p>	<p>Existing Control Measures</p> <p>Avoid lone or out-of-hours working</p>
<p>Risk Level</p> <p>With Existing Controls:</p> <p><u>Risk Level</u> C- Medium / Moderate</p>	

Appendix C: MATLAB Codes Used to Analyse Laser Tomography Images and PIV Data

C1: This code was written to enable visualisation of the tomographic images of the flame kernel in pseudo-coloured form.

```

clc
close all
clear all
tic
folderpath='E:\21032019_PIV\LAM_TIF\New folder';% Current working folderpath (no more than
2 subfolder)
imagetype='*.TIF';% type of image in the folder
file=fullfile(folderpath,imagetype);
directory=dir(file);
Numimage=numel(directory);
delt=round([1 51 167.67:166.67:(1+166.67*10)]);

for k=1:Numimage
ff= fullfile(folderpath,directory(k).name);
Raw_image_x=imread(ff);
M=double(rgb2gray(Raw_image_x));
% M1=M(1:end-25,:);
M2=flip(M,1);
% M2=imrotate(M,-180);
% M2=flip(M1,2);
% M1=imcrop(M,[50 100 300 300]);
[ax,ay]=size(M2);

a=52e-3;
x=1:ax;
y=1:ay;
[X,Y]=meshgrid(y,x);

max_pix=max(max((M2)));
%max_pix=1;
min_pix=0;

if k<10, continue; end

figure(1),
% hold on;
subplot(1,3,k-9),
pcolor (a*X,a*Y,(M2)), axis on, %axis equal;
shading flat, caxis([min_pix max_pix]),title([num2str(delt(k)), '  $\mu$ s']),%colorbar ('vert')
% hold on;
% imtool(figure(),[num2str(delt(k)) '.jpeg'],'.jpeg');
end

```

C2: This code was written to extract the kernel edges from the Laser tomography image and to obtain the flame tip locations and propagation rate

```

clear all
close all
clc

tic
folderpath='E:\New folder\Kernel Xtics\Laminar\New folder';% Current working folderpath (no
more than 2 subfolder)
imagetype='*.TIF';% type of image in the folder
file=fullfile(folderpath,imagetype);
directory=dir(file);
Numimage=numel(directory);

delt=round([51:166.67:166.67*12]);

for k=1:length(delt)
    % for k=1:Numimage
    ff= fullfile(folderpath,directory(k).name);
    Mask_image_x=imread(ff);
    M=im2double(Mask_image_x);
    M1=M(250:600,100:250);
    % M4=flip(M1,1);
    M2=imcomplement(M1);
    M3=edge(M2,'canny',0.0001);

    DF=diff(sum(M3'));

    [peaks, locs]=findpeaks(DF);
    DF1=diff(M3(min(locs),:));
    findpeaks(diff(M3(min(locs),:)))
    % %Yt(fi,di)=min(find(diff(sum(KP'))==max(peaks))); % higher point on the kernel
    Yt(k,:)=min(locs);
    % indexxt=find(M3(min(locs)+1,:)==1);
    % Xt=ceil(mean(indexxt));
    [peaks1, locs1]=findpeaks(-1*DF);
    % Yb(fi,di)=max(find(-1*diff(sum(KP'))==max(peaks1))); % lowest point on the kernel
    Yb(k,:)=max(locs1);

    % if k>3, stop; end
    %
    % figure(1),
    % subplot(1,3,k), imshow(M3), title([num2str(delt(k)), ' μs']),axis on, %axis equal;
    % % subplot(1,3,k), imshow(edgematrix), title([num2str(delt(k)), ' μs']), colorbar verti, axis
    equal;
    %

end

Yt1=351-Yt;
Yb1=351-Yb;
Y=Yt1-Yb1;

```

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```
figure(100),plot(delt,52e-3.*Yt1,'ko-'); hold on; plot((delt),52e-3.*Yb1,'r--*'); xlabel('time,  $\mu$ s'),  
ylabel('location, mm');  
legend('downstream location', 'upstream location');  
figure(101),scatter(delt,52e-3.*Y,'ko'); xlabel('time,  $\mu$ s'), ylabel('location, mm');
```

C3: This code was written to visualize the flow field velocity from the PIV data

```
close all  
clc  
  
%%%%%%Reads Multiple excel file%%%%%%%%  
folderpath='E:\21032019_PIV\LAM_VEC';%14F11A VECTOR FILES  
filetype='*.xls';  
file=fullfile(folderpath,filetype);  
directory=dir(file);  
N=numel(directory);  
u=cell({});  
v=cell({});  
n=N;  
% data{k}=cell({});  
us=[];  
vs=[]  
  
ux=[];  
vy=[]  
  
for k=1:n  
    data{k}=xlsread(fullfile(folderpath,directory(k).name));  
    cv=folderpath;directory(k).name  
    x=data{k}(1:end,1);  
    y=data{k}(1:end,2);  
    ux=[ux,x];  
    vy=[vy,y];  
    % if k==1  
    u=data{k}(1:end,3);  
    v=data{k}(1:end,4);  
    us=[us,u];  
    vs=[vs,v];  
  
    rgx=min(ux(:,k)):0.5:max(ux(:,k));  
    rgy=min(vy(:,k)):0.5:max(vy(:,k));  
    % grd=griddata(rgx,rgy,ux(:,k),vy(:,k),us(:,k),vs(:,k));  
  
    xmin=min(ux(:,k));  
    xmax=max(ux(:,k));  
  
    ymin=min(vy(:,k));  
    ymax=max(vy(:,k));  
  
    y1=ymin:0.05:ymax;  
    x1=xmin:0.05:xmax;  
  
    [x2d,y2d] = meshgrid(x1,y1);  
    velu = griddata(ux(:,k),vy(:,k),us(:,k),x2d,y2d);  
    velv = griddata(ux(:,k),vy(:,k),vs(:,k),x2d,y2d);  
    velmag = griddata(ux(:,k),vy(:,k),sqrt(us(:,k).^2+vs(:,k).^2),x2d,y2d);
```

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```
figure()
pcolor(x2d,y2d,velv), axis image, colormap jet, colorbar verti, shading flat
hold on
quiver(ux(:,k),vy(:,k),us(:,k),vs(:,k),1.5,'filled','k'),set(gca,'color',[1 1 1]);

end
%% =====
```

Appendix D: MATLAB Code Used to Analyse Chemiluminescence Data

This MATLAB code visualises the pseudo-colour chemiluminescence image of the kernel, extracts image edges and outputs OH* kernel tip locations and propagation rates

```
clc;
clear all;
close all;
CurrentPath=pwd;
%% =====
prefix_FolderName=['aS29Test'];
startF=15; endF=15; %starting and ending folder number
%outfile name

%delta t - time delay from laser ignition, for all cases following delay
%used; the values are in microseconds

delt=(150:50:500 600:100:1000 1500:500:2000 3000:500:5000 6000:1000:10000)];
delts= delt*1e-6; % converted to seconds

for fi=startF:endF
    FolderName=[prefix_FolderName num2str(fi)];
    cd(FolderName) % change the working folder
    pwd
    dirinfo=dir('a*.SPE');
    name_list={dirinfo.name};
    for di=1:length(delt) %#ok<ALIGN>
        %
        for fii=1:length(name_list)
            if contains(char(strcat(name_list(fii)),'T' num2str(delt(di)))), filename=strcat(name_list(fii));
            end
            if fii==18, continue; end
        end %fii % getting filename for loading
        filename=char(filename);
        filename1=filename(1:end-4); % name without suffix

        a=readSPE(char(filename)); % load SPE file into matrix
        mean_a=mean(a,3); % mean image from the raw chemi images
        [mx, my]=size(mean_a);

        %region of interest definition
        %top left, bottom right
```

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```
t=167; b=mx-45; sr=320;sl=20;
% t=67; b=mx-45; sr=320;sl=20;

%
% ROI=imcrop(mean_a,[1 167 300 300]);
ROI=mean_a(t:b,sl:sr);
% ROI=mean_a(1:my,1:mx);

TL=20;
aLevel=multithresh(ROI,TL); % multilevel threshold, here TL=1
seg_a=imquantize(ROI,aLevel);
%PV=seg_a-1; % progress variable based on thresholding.. requires modification to include inside
area of kernel
if delt(di)<8000, customlevel=4; else, customlevel=4; end
M1=imbinarize(ROI,aLevel(customlevel));
filtersize=7;
PV=medfilt2(M1,[filtersize filtersize]);%% progress variable based on thresholding.. requires
modification to include inside area of kernel

TMa=ROI.*(seg_a-1); %this will generate a thresholded mean iamge
TMa(TMa<0)=0;
% figpv=figure(2),imagesc(PV), colorbar hori, axis equal
KP=edge(PV); %edge on PV gives kernel perimeter
% figkp=figure(3),imagesc(KP), colorbar hori, axis equal

DF=diff(sum(KP'));
%
[peaks, locs]=findpeaks(DF);
DF1=diff(KP(min(locs),:));
findpeaks(diff(KP(min(locs),:)))
% Yt(fi,di)=min(find(diff(sum(KP'))==max(peaks))); % higher point on the kernel
Yt(fi,di)=min(locs);
indexxt=find(KP(min(locs)+1,:)==1);
Xt(fi,di)=ceil(mean(indexxt));
[peaks1, locs1]=findpeaks(-1*DF);
% Yb(fi,di)=max(find(-1*diff(sum(KP'))==max(peaks1))); % lowest point on the kernel
Yb(fi,di)=max(locs1);
indexxb=find(KP(max(locs),:)==1);
Xb(fi,di)=ceil(mean(indexxb));

Itotal(fi,di)=sum(sum(ROI));
Imax(fi,di)=max(max(ROI));

edgematrix=zeros(size(KP));
edgematrix(Yt(fi,di),:)=1;
edgematrix(Yb(fi,di),:)=1;

% figure (1),
% subplot(4,4,di), imagesc(ROI), title([num2str(delt(di)), ' μs']), colormap jet, axis equal; axis off
% subplot(4,4,di), imagesc(ROI), title([num2str(delt(di)), ' μs']), colormap jet, %axis equal, %axis
off,
% colorbar verti, set(gca, 'FontSize', 18),set(gca, 'FontSize', 2),
% subplot(3,3,di), imagesc(M1), title([num2str(delt(di)), ' μs']), colorbar verti, axis equal;
% subplot(3,3,di), imagesc(PV), title([num2str(delt(di)), ' μs']), colorbar verti, axis equal;
```

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```
% subplot(3,3,di), imagesc(KP), title([num2str(delt(di)), ' µs']), colorbar verti, axis equal;
% subplot(4,1,di), imagesc(KP+edgematrix), title([num2str(delt(di)), ' µs']), colorbar verti, axis
equal;
% pause(0.01);

end %% di, delay loop

%
cd(CurrentPath) % change it back to main folder
% %
% path=('D:\Flame_kernel_chemiluminescence\Sunny\Chapter6\delt.mat');
% save(path,'delt');

Xt1=20+Xt(fi,:);
Yt1=345-Yt(fi,:);
Xb1=20+Xb(fi,:);
Yb1=345-Yb(fi,:);
XYtop=[Xt1(:),Yt1(:)];
XYbot=[Xb1(:),Yb1(:)];

% Xt1=20+Xt; Yt1=445-Yt;
% Xb1=20+Xb; Yb1=445-Yb;
% XYtop=[Xt1(:),Yt1(:)];
% XYbot=[Xb1(:),Yb1(:)];

path3a=['E:\Flame_kernel_chemiluminescence\Sunny\Chapter6\', [prefix_FolderName
num2str(fi)], '_XYtop.mat'];
save(path3a,'XYtop');
path3b=['E:\Flame_kernel_chemiluminescence\Sunny\Chapter6\', [prefix_FolderName
num2str(fi)], '_XYbot.mat'];
save(path3b,'XYbot');
figure (3), scatter(Xt1,Yt1,'g*'), xlim([150 200]), grid on, grid minor; hold on,
scatter((Xb1),(Yb1),'r*'), xlim([150 200]), grid on, grid minor;
hold on, title('scatter plot of flame tip location'), xlabel('X, pixel'), ylabel('Y, pixel'),
legend('downstream location', 'upstream location')

figure (4), plot(delt,Itotal(fi,:),'r*'), xlabel('time, µs'), ylabel('integrated intensity, au'), grid on, grid
minor, title('integrated intensity vs time');
path4=['E:\Flame_kernel_chemiluminescence\Sunny\Chapter6\', [prefix_FolderName
num2str(fi)], '_Itotal.mat'];
save(path4,'Itotal');

figure (5), plot((delt),Imax(fi,:),'r*'), xlabel('time, µs'), ylabel('peak intensity, au'), grid on, grid
minor, title('peak intensity vs time');
path5=['E:\Flame_kernel_chemiluminescence\Sunny\Chapter6\', [prefix_FolderName
num2str(fi)], '_Imax.mat'];
save(path5,'Imax');

figure (6), scatter(delt,185e-3.*Yt1,'go'); hold on; scatter((delt),185e-3.*Yb1,'r^'); xlabel('time, µs'),
ylabel('location, mm');
legend('downstream location', 'upstream location');
path6a=['E:\Flame_kernel_chemiluminescence\Sunny\Chapter6\', [prefix_FolderName
num2str(fi)], '_Yt1.mat'];
save(path6a,'Yt1');
```

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```
path6b=['E:\Flame_kernel_chemiluminescence\Sunny\Chapter6\', [prefix_FolderName
num2str(fi)], '_Yb1.mat'];
save(path6b,'Yb1');

for x=1:1
Yt01=185e-6*Yt1(x,:);
Yb01=185e-6*Yb1(x,:);

Ut=diff(Yt01)./diff(delts);
Ub=diff(Yb01)./diff(delts);

figure (7), plot(delts(2:length(delts)),Ut,'go'); hold on
plot(delts(2:length(delts)),Ub,'r^'); hold on;
title('Growth rate of Torroidal flame'), xlabel('time, sec'), ylabel('flame speed, m/s'),
legend('downstream flame', 'upstream flame')
path7a=['E:\Flame_kernel_chemiluminescence\Sunny\Chapter6\', [prefix_FolderName
num2str(fi)], '_Ut.mat'];
save(path7a,'Ut');
path7b=['E:\Flame_kernel_chemiluminescence\Sunny\Chapter6\', [prefix_FolderName
num2str(fi)], '_Ub.mat'];
save(path7b,'Ub');

end %x

end %fi folder loop
```

Appendix E: Sensitivity Analysis

Table 0.1: Pulse Energy effect on the Kernel development parameters

Parameter	Pulse Energy (mJ)			
	32	64 (100%)	96 (200%)	128 (300%)
Dimension: W @ 1 ms (mm)	13.13	16.83 (28%)	17.94 (37%)	18.13 (38%)
Dimension: W @ 10 ms (mm)	38.48	37.00	37.18	37.37
Growth rate: S_{y+} @ 0.45 ms	3.70	5.55 (50%)	5.55 (50%)	5.55 (50%)
Growth rate: S_{y+} @ 1.0 ms	3.70	3.70	3.70	3.70
Minimum integ OH* intensity (AU)	4.43E+07	3.68E+07	3.90E+07	4.24E+07
Time @ min integ OH* intensity (μ s)	7	7	7	7
Maximum integrated OH* intensity (AU)	5.80E+07	4.37E+07 (-25%)	4.45E+07 (-23%)	5.69E+07 (-2%)
Min peak intensity (AU)	1314	1084 (-18%)	1071 (-19%)	1090 (-17%)
Time @ min peak intensity (ms)	1.5	2	3	3

Table 0.2: Effects of Equivalence ratio on Kernel development parameters (Laminar flow)

Parameter	Equivalence Ratio (Laminar)		
	1.00	0.90	0.80
Y_+ @7 ms (mm)	28.405	25.815 (9%)	23.225 (18%)
Steady state S_{y+} (m/s)	3.33	2.96 (-11%)	2.59 (-22%)
minimum peak OH* intensity (AU)	1.31E+03	1.07E+03 (-18%)	8.11E+02 (-38%)
time at minimum peak OH* intensity (ms)	1.5	1.5	2
maximum integrated OH* intensity (AU)	5.80E+07	5.30E+07 (-9%)	4.66E+07 (-20%)
minimum integ OH* intensity (AU)	4.43E+07	4.79E+07	4.42E+07
time at minimum integrated OH* intensity (μ s)	200	200	450

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Table 0.3: Effects of Equivalence ratio on Kernel development parameters (Turbulent flow)

Parameter	Equivalence Ratio (Turb)		
	1.00	0.90	0.80
Y_+ @7 ms (mm)	51.53	48.385 (6%)	48.015 (7%)
Steady state S_{y+} (m/s)	2.14	2.16 (1%)	1.43 (33%)
minimum peak OH* intensity (AU)	1.21E+03	9.39E+02 (22%)	7.55E+02 (38%)
time at minimum peak OH* intensity (μ s)	1	3	3.5
maximum integrated OH* intensity (AU)	7.71E+07	6.93E+07 (10%)	6.68E+07 (13%)
minimum integ OH* intensity (AU)	6.08E+07	6.26E+07	6.32E+07
time at minimum integrated OH* intensity (μ s)	350	350	450

Table 0.4: Effect of the fuel on the Kernel development parameters (Constant AFT)

Parameter	T=2226 K			T=1234 K		
	CH ₄	C ₃ H ₈	C ₂ H ₄	CH ₄	C ₃ H ₈	C ₂ H ₄
Maximum Y_- (mm)	12.295 (0%)	6.745 (-45%)	9.89 (-20%)	10.26 (0%)	-0.10 (-101%)	7.115 (-42%)
Steady state S_{y+}/S_L	1.1275 (0%)	0.7148 (-37%)	0.8925 (-21%)	1.0269 (0%)	0.5616 (45%)	0.7650 (-26%)
minimum peak OH* intensity (AU)	1.31E+03 (0%)	1.04E+03 (-21%)	1.51E+03 (15%)	1.07E+03 (0%)	6.42E+02 (-40%)	1.16E+03 (+8%)
time at minimum peak OH* intensity (ms)	1.5	2	1			
maximum integrated OH* intensity (AU)	5.80E+07 (0%)	4.92E+07 (-15%)	6.12E+07 (+6%)	52449275 (0%)	44441798 (-15%)	56525861 (+8%)
minimum integ OH* intensity (AU)	4.43E+07	4.10E+07	5.13E+07	47913172	42883432	49973437
time at minimum integrated OH* intensity (μ s)	200	250	500	200	350	400

Table 0.5: Effect of the fuel on the Kernel development parameters (Constant LfV)

Parameter	LFV=41 cm/s	LFV=29 cm/s
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Appendices

	CH4	C3H8	C2H4	CH4	C3H8	C2H4
Y_+ (mm) @ 7ms	31.18	29.33 (-6%)	33.03 (+6%)	23.225	17.86 (-23%)	20.265 (-13%)
Steady state S_{y+}/S_L	1.1275 45	0.8744 56 (-22%)	1.3041 03 (16%)	1.449843 26	0.824200 3 (-43%)	1.2131 81 (-16%)
minimum peak OH* intensity (AU)	1.31E+ 03	1.36E+ 03 (+4%)	1.80E+ 03 (32%)	8.11E+0 2	5.34E+0 2 (-34%)	7.26E+ 02 (-11%)
time at minimum peak OH* intensity (μ s)	1.5	1.5	1	2	4.5	4
maximum integrated OH* intensity (AU)	5.80E+ 07	5.01E+ 07 (-14%)	6.66E+ 07 (33%)	4650484 3.8	3689688 3.2 (-20%)	554397 66 (50%)
minimum integ OH* intensity (AU)	4.43E+ 07	3.85E+ 07	5.25E+ 07	4424647 8.4	3739389 7.2	495093 89
time at minimum integrated OH* intensity (μ s)	200	250	250	450	450	700