Direct Numerical Simulation of Lean Premixed Turbulent Flames at High Karlovitz Numbers under Elevated Pressures

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This thesis is submitted for the degree of
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To my mother.
Declaration

I hereby declare that the work presented in this thesis is my own and that I have correctly acknowledged the work of others. This submission is in accordance with University College London and the Department of Mechanical Engineering on proper academic conduct.

Signature:

Date:
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Abstract

Lean premixed combustion is a promising strategy for the next generation of gas turbines, which is characterised by low pollutant emissions and high combustion efficiency. However, flame quenching and combustion instability arising from this technique could increase operating cost and decrease operating efficiency. The fundamentals behind these problems are not yet clarified. Therefore, it is essential to study and understand fundamental combustion phenomena of lean premixed flames in conditions relevant to practical combustion devices, which will promote the development of turbulent flame models and help to get full advantages of this technique.

With the availability of increasingly powerful supercomputers, direct numerical simulation (DNS) of turbulent reacting flow has become feasible and affordable. This thesis investigates lean premixed turbulent H₂/air flames at high Karlovitz (Ka) numbers under elevated pressures by using DNS with multi-step chemistry. The effects of the Karlovitz number, pressure, equivalence ratio and integral length scale on flame structures and chemical pathways are examined qualitatively and quantitatively.

It is found that the relative probability of positive curvature to negative curvature is insensitive to Ka but sensitive to pressure and integral length scale ($l_i$). On flame fronts, the local heat release rates in regions with high-positive curvatures are higher than those in regions with high-negative curvatures when conditioned on the same H₂ consumption rate, whereas this phenomenon is getting weaker with decreasing Ka and increasing pressure. As pressure increases, the flame speed and thickness ($\delta_L$) decreases, and the reaction zone moves to regions with higher values of progress variable. Moreover, the thickness of the inner layer conditioned on the laminar flame thickness becomes smaller under elevated pressures, which results in a lower probability of
finding high curvatures in the high-pressure flames with a fixed Ka. Under conditions relevant to gas turbines, the heat release rate and scaled reaction zone thickness ($\delta_f/\delta_L$) increase with increasing equivalence ratio. However, flames demonstrate similar topological structures of flame fronts when Ka is fixed. Trenches of local equivalence ratio ($\varphi_L$) with small gradients are observed in concave structures outside the reaction zone, while $\varphi_L$ plateaus with large gradients are observed in convex structures inside the reaction zone. When the integral length scale is smaller than the thickness of the corresponding laminar flame, turbulence is unable to stretch and interrupt the reaction zone and the flame presents laminar flamelet characteristics. However, the distributions of curvature and tangential strain rate are comparable with those in the same Ka flames with $l_t/\delta_L \geq 1.0$. It is also found that keeping constant $l_t/\delta_L$ ratio and Ka could isolate the effects of pressure on flame front structures. The turbulent flame with unity $l_t/\delta_L$ ratio could capture the main features of heat release as those in flames with higher $l_t/\delta_L$ ratios. Considering the chemical process, pressure could significantly modify the chemical pathways in both laminar and turbulent flames, and the effects are more significant than those of the Karlovitz number and integral length scale. Due to the combined effects of radical fractions and reaction rate constants, the local heat release is changed in different temperature windows when the mixture equivalence ratio varies.
Impact Statement

Lean premixed combustion is a promising strategy for NO\textsubscript{x} emission reduction and combustion efficiency improvement. It has been employed in industrial combustion devices, including international combustion engines, gas turbines and furnaces. However, insufficient understanding of this technology increases operating cost, decreases operating efficiency and limits its application. This work extends the ongoing DNS studies to lean premixed turbulent flames at high Karlovitz numbers (Ka) and under elevated pressures. The key impacts of this study are:

- The effects of Karlovitz number, pressure, equivalence ratio and integral length scale on flame structures and chemical pathways in lean premixed H\textsubscript{2}/air flames are investigated comprehensively, which contributes to our knowledge of high-Ka flames under elevated pressures.
- The simulations are performed under conditions relevant to practical combustion engines. Physical insight into complex combustion processes has been presented. It helps to understand combustion phenomena in industrial devices and supplies a guideline for the design of combustion engines.
- A precise combustion model is vital in the design of industrial combustion devices. The DNS database could be used to validate and develop combustion models for engineering applications at high Ka under elevated pressures.
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Nomenclature

\( A_i \)  pre-exponential factor of Arrhenius equation [cm\(^3\)/g·s]

\( a_{ki} \)  contribution rate of species \( k \) in Three-body reactions

\( a_T \)  tangential strain rate [s\(^{-1}\)]

\( C_{hr,i} \)  global heat release contribution of reaction \( i \)

\( C_{local,i} \)  local heat release contribution of reaction \( i \)

\( c \)  progress variable

\( c_p \)  heat capacity at a constant pressure [erg/K]

\( D_{jk} \)  binary diffusion coefficient [cm\(^2\)/s]

\( E_i \)  activation energy [erg/mol]

\( E(\kappa_n) \)  kinetic energy density [cm\(^3\)/s\(^2\)]

\( F_{visc} \)  viscous force [kg/m\(^2\)·s\(^{-2}\)]

\( H_k^0 \)  enthalpy of species \( k \) [erg/mol]

\( HR_{r,i} \)  relative heat release rate of reaction \( i \)

\( h_k \)  enthalpy [erg/mol]

\( J_k \)  diffusion flux of species \( k \) [kg/m\(^2\)·s]

\( K \)  flame stretch rate [s\(^{-1}\)]

\( K_0 \)  limit of low-pressure reaction rate constant [s\(^{-1}\)]

\( K_{\infty} \)  limit of high-pressure reaction rate constant [s\(^{-1}\)]

\( K_a \)  Karlovitz number

\( K_{fi} \)  forward reaction rate constant of reaction \( i \) [s\(^{-1}\)]

\( K_{pi} \)  equilibrium constant

\( K_{ri} \)  reverse reaction rate constant of reaction \( i \) [s\(^{-1}\)]
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$l_\delta$</td>
<td>inner layer thickness</td>
<td>[cm]</td>
</tr>
<tr>
<td>$l_c$</td>
<td>Taylor length scale</td>
<td>[cm]</td>
</tr>
<tr>
<td>$l_t$</td>
<td>integral length scale</td>
<td>[cm]</td>
</tr>
<tr>
<td>$\bar{M}$</td>
<td>molecular weight of mixture</td>
<td>[g/mol]</td>
</tr>
<tr>
<td>$M_k$</td>
<td>molecular weight of species $k$</td>
<td>[g/mol]</td>
</tr>
<tr>
<td>$P$</td>
<td>pressure</td>
<td>[atm]</td>
</tr>
<tr>
<td>$q$</td>
<td>heat flux</td>
<td>[erg/cm$^3$·s]</td>
</tr>
<tr>
<td>$R$</td>
<td>universal gas constant</td>
<td>[erg/mol·K]</td>
</tr>
<tr>
<td>Re</td>
<td>Reynolds number</td>
<td></td>
</tr>
<tr>
<td>$S$</td>
<td>trace-less rate of strain tensor</td>
<td>[s$^{-1}$]</td>
</tr>
<tr>
<td>$S_d$</td>
<td>displacement speed</td>
<td>[cm/s]</td>
</tr>
<tr>
<td>$S_k^0$</td>
<td>entropy of species $k$</td>
<td>[erg/K]</td>
</tr>
<tr>
<td>$S_L$</td>
<td>laminar flame velocity</td>
<td>[cm/s]</td>
</tr>
<tr>
<td>$T$</td>
<td>temperature</td>
<td>[K]</td>
</tr>
<tr>
<td>$T_b$</td>
<td>burnt gas temperature</td>
<td>[K]</td>
</tr>
<tr>
<td>$T_u$</td>
<td>unburnt gas temperature</td>
<td>[K]</td>
</tr>
<tr>
<td>$t_F$</td>
<td>chemical time scale</td>
<td>[s]</td>
</tr>
<tr>
<td>$t_\eta$</td>
<td>Kolmogorov time scale</td>
<td>[cm]</td>
</tr>
<tr>
<td>$U$</td>
<td>velocity</td>
<td>[cm/s]</td>
</tr>
<tr>
<td>$\bar{u}$</td>
<td>time-averaged velocity</td>
<td>[cm/s]</td>
</tr>
<tr>
<td>$u'$</td>
<td>root-mean-square (RMS) turbulent velocity</td>
<td>[cm/s]</td>
</tr>
<tr>
<td>$u_k$</td>
<td>Kolmogorov characteristic velocity</td>
<td>[cm/s]</td>
</tr>
<tr>
<td>$u_i$</td>
<td>velocity component in $i$-direction</td>
<td>[cm/s]</td>
</tr>
<tr>
<td>$V_k$</td>
<td>diffusion velocity of species $k$</td>
<td>[cm/s]</td>
</tr>
</tbody>
</table>
$X_k$  mole fraction of species $k$

$Y_k$  mass fraction of species $k$

**Greek Symbols**

$\beta_i$  temperature exponent of the Arrhenius equation

$\delta_{ij}$  Kronecker delta

$\delta_L$  laminar flame thickness [cm]

$\varepsilon$  turbulence dissipation rate [cm$^2$/s$^3$]

$\eta$  Kolmogorov length scale [cm]

$\kappa$  curvature [cm$^{-1}$]

$\kappa_n$  wavenumber [cm$^{-1}$]

$\lambda$  thermal conductivity [erg/s·cm·K]

$\nu$  kinematic viscosity [cm$^2$/s]

$\rho$  mixture density [g/cm$^3$]

$\tau_l$  large eddy turn-over time [s]

$\nu_{ki}^f$  forward stoichiometric coefficient for species $k$ in reaction $j$

$\nu_{ki}^r$  reverse stoichiometric coefficient for species $k$ in reaction $j$

$\phi$  equivalence ratio

$\phi_L$  local equivalence ratio

$\Omega_g$  antisymmetric part of gradient velocity tensor [s$^{-1}$]

$\dot{\omega}$  reaction rate of species $k$ [mol/cm$^3$·s]

**Other Symbols**

$(F/A)_{stoic}$  stoichiometric air-fuel ratio
[M] overall contribution of different species in Three-body reactions

**Abbreviations**

CDF cumulative distribution function

CFD computational fluid dynamics

DNS direct numerical simulation

FCR fuel consumption rate [mol/cm$^3$·s]

HRR heat release rate [erg/cm$^3$·s]

LES large eddy simulation

LSODE Livermore Solver for Ordinary Differential Equations

NSCBC Navier-Stokes Characteristic Boundary Conditions

PDF probability density function

PIV particle image velocimetry

PLIF planar laser-induced fluorescence

RANS Reynolds-averaged Navier-Stokes

RK3-2N third-order Runge-Kutta

RST rayleigh scattering thermometry

SDF surface density function
Chapter 1

Introduction

This chapter presents the background and motivations of this research. Moreover, the structure and objectives of this thesis are listed.

1.1 Background

The relationship between human beings and combustion can be dated back to millions of years ago when humans started to use fire to cook food, keep warm and scare wild animals away. The fire ignited human evolution, and the control of fire was then closely integrated into the evolutionary history of human [1]. The direct benefit of using fire for cooking was the enhanced consumption of proteins which contributes to a significant physiological development into Homo ergaster and Homo erectus [2]. Gradually, the brain size of Homo increased and thus promoted humans to make and use stone tools. It is fair to say that the advancements in fire utilisation reflect the evolution of human brains. Figure 1.1 shows a painting by British artist Peter Jackson that a Stone Age mother and child are making a fire. ‘Drill fire making’ was a significant but straightforward technology in human evolution, which helped humans to get rid of the dependence on natural fire. Moreover, the use of fire allowed humans to protect themselves from predators and warm themselves in cold weathers [3], which
significantly expanded the living space for human beings and extended the length of lifetime.


Ever since the discovery of the role of oxygen in combustion by Antoine Lavoisier in the 18th century, combustion science has turned to a new chapter. The advent of combustion engines and power plants stimulated scientific researches in combustion theory and application. From the mid-1970s, the rapid development in mathematical analysis and experimental techniques have significantly advanced the modern combustion science [4]. Combustion technology even helps people to explore the mystery of the universe. On 12 August 2018, NASA launched Parker Solar Probe, making humans’ first touch to the sun. Figure 1.2 shows the powerful propulsion system. This launcher is equipped with a solid-fuel upper stage that will supply abundant energy
to allow the spacecraft to fall into the inner solar system from the earth [5]. There is no
doubt that humans’ understanding and application of combustion under extreme
conditions have reached a new level.

Fig. 1.2 NASA launched Parker Solar Probe to explore the Sun. Digital image. Source:

As mentioned by Pausas and Keeley [6], on one hand, the fire has strong
evolutionary consequences for human; on the other hand, human activities have
inevitably affected the ecosystems and its sustainability. Combustion supplies energy
to maintain the normal production and living of humans. In the past decades,
combustion of fossil fuels is the main source of energy in industry, transport and
residential area. According to the latest statistics [7], oil, coal and natural gas contribute
around 85% of the world’s total energy supply. As can be seen from Fig. 1.3, the share
of coal had been slightly increased, and the percentage of oil had been dramatically
decreased. Furthermore, the percentage of natural gas had been increased in the past 45
years. Unfortunately, the combustion of fossil fuels, especially coal and oil, produces
environmental pollutants such as NO\textsubscript{x}, SO\textsubscript{x} and particulate matter. It should also be noted that the world total primary energy supply dramatically increased from 5523 Mtoe to 13761 Mtoe from the year 1971 to 2016 due to the population growth and economic development [7], which has raised the social concerns about energy sufficiency and environmental pollution. Moreover, global warming aggravated by CO\textsubscript{2} emissions from the combustion of fossil fuels is another scientific concern in the 21\textsuperscript{st} century [8, 9].

![Fig. 1.3 Total primary energy supplies in 1971 and 2016.](image)

1.2 Motivations

Our domestic life, transportation system and industrial processes all rely heavily on combustion. However, the downside issues, e.g. environmental pollution and ecological damage, caused by combustion cannot be ignored. Moreover, the resource of fossil fuels is limited, and it cannot be replaced in a short time. As energy security and environmental quality are becoming global issues, it is crucial to enrich our
knowledge of combustion phenomena, which will help to increase combustion efficiency and optimise energy structures.

![Diagram of combustion phenomena](image)

**Fig. 1.4** Sketches of flame structure for (a) premixed combustion and (b) non-premixed combustion.

Generally, combustion can be categorised as either premixed or non-premixed combustion according to the mixing state of fuel and oxidiser [10, 11]. In premixed combustion, the fuel and oxidiser are fully mixed at molecular level prior to combustion. Non-premixed flame is also known as diffusion flame where the oxidiser and fuel are initially separated, and the diffusion driven by concentration gradients promotes the mixing of reactants. As a result, the reactions happen at the interface between the fuel and oxidiser. The general flame structures of premixed and non-premixed flames are given in Fig. 1.4. It is noted that, for premixed flames, the temperature of the premixed gas will rise due to the intense heat release in the reaction zone and the burned products are characterised by high temperatures. For non-premixed flames, the air and fuel are separated by the reaction zone where the peak temperature occurs. Furthermore, there is also a process called partially premixed combustion, in which the reactants are partially mixed prior to combustion.
In practical combustion, non-premixed and partially premixed combustions typically occur in diesel engines and furnaces whereas premixed combustion is found in homogeneous compression charge ignition engine and lean-burn gas turbine [12]. Generally, non-premixed combustion has a wider application due to its strong stability over an extensive range of operating conditions. However, one fatal defect of this technology is the high level of NO\textsubscript{x} emission, which conflicts tightening emission regulations [13]. For modern combustion devices, emissions reductions are considered as a fundamental design constraint and a principal aspect [14]. Lean premixed combustion has the characteristic of low flame temperature, which supplies a promising strategy for emission control of NO\textsubscript{x} and soot. However, the understanding of lean premixed combustion is still insufficient, and this limits the application of lean premixed combustion in commercial devices [13]. Further exploration of this strategy is vital to get full advantages of this technique. As such, it is essential to study and understand fundamental combustion phenomena of lean premixed flames in conditions relevant to practical combustion devices, which will promote the validation and development of turbulent flame models for engineering applications.

At present, there are three major techniques to study the fundamental phenomena of premixed combustion: theoretical analysis, experimental measurement and computational simulation. In theoretical studies, the major work is focusing on formula deduction, which is restricted to simple geometries and linear problems. In experimental studies, spherical flame, jet flame and counterflow flame are commonly measured [15-20]. In order to measure the flame properties accurately, several non-contact measuring methods need to be adopted. For example, high-speed camera is used to capture flame propagation images [19]; Rayleigh scattering thermometry (RST) is
1.2 Motivations

used to detect temperature field [21]; planar laser-induced fluorescence (PLIF) is
applied to investigate the fundamentals of the flow dynamics and flame front structures
[22]; particle image velocimetry (PIV) is applied to measure the flow field [23].
However, these measuring systems are usually complicated and expensive. The
accurate description of the interaction between turbulence and combustion is
challenging, especially at high turbulence intensity and under elevated pressure.
Fortunately, with the availability of increasingly powerful computers, computational
fluid dynamics (CFD), which can solve highly non-linear problems, is becoming an
essential tool in the design of aerodynamical systems. Computational simulation of
combustion has become an essential tool for both engineers and researchers, which
improves the efficiency of industrial production and brings essential improvements to
our understanding of combustion phenomena [24, 25].

For practical combustion devices, the operation conditions are usually characterised
by high turbulence intensity and high pressures [26, 27], which requires high spatial
and temporal resolutions for both experimental and numerical studies. However, it is
pretty difficult to set up experiments and numerical cases under conditions relevant to
practical combustion, and this limits our understanding in turbulent combustion at
extreme operation conditions. In recent years, computational simulation of combustion
is becoming the focus of research, and the key issues of simulation are combustion and
turbulence models. Good models are vital for accurate predictive simulations.
Nevertheless, most of the combustion and turbulence models are verified under low-
intensity turbulence and the atmospheric pressure. As such, it is essential to enrich our
knowledge of combustion at high turbulence intensities and under high pressures, so
that combustion and turbulence models could be improved to achieve a better prediction of practical combustion.

1.3 Simulation strategies

There are various methods with different application areas to compute reacting flows. The three most widely used approaches in combustion simulation are Reynolds-averaged Navier-Stokes (RANS), large eddy simulation (LES) and direct numerical simulation (DNS) [25]. Table 1.1 lists the key properties of these strategies in combustion simulation. Figure 1.5 shows the schematic diagram of the turbulent energy spectrum [11, 28] with a description of resolved turbulent scales for different simulation approaches.

Table 1.1 List of properties for different simulation strategies

<table>
<thead>
<tr>
<th>Numerical Strategy</th>
<th>RANS</th>
<th>LES</th>
<th>DNS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mesh resolution</td>
<td>Low</td>
<td>Moderate</td>
<td>High</td>
</tr>
<tr>
<td>Resolved turbulent scales</td>
<td>Mean flow field</td>
<td>Large scales</td>
<td>All scales</td>
</tr>
<tr>
<td>Simulation configurations</td>
<td>Complex</td>
<td>Moderate</td>
<td>Simple</td>
</tr>
<tr>
<td>Computational cost</td>
<td>Low</td>
<td>Moderate</td>
<td>High</td>
</tr>
<tr>
<td>Application area</td>
<td>Industry</td>
<td>Research&amp;Industry</td>
<td>Research</td>
</tr>
</tbody>
</table>

In turbulent energy spectrum, the kinetic energy density is expressed as [11]:

$$E(\kappa_n) = C \varepsilon^{2/3} \kappa_n^{-5/3}$$

(1.1)

where $C = 1.5$ and $\varepsilon$ is the turbulence dissipation rate. $\kappa_n$ is the wavenumber and inversely proportional to the eddy size $l_n$ as:
1.3 Simulation strategies

\[ \kappa_n \propto l_n^{-1} \] (1.2)

In Fig. 1.5, large eddies (or small wavenumbers) contain most of the turbulent kinetic energy (TKE), and this range is called energy-containing range. Small eddies (or large wavenumbers, corresponding to the dissipation range) do not contribute TKE, but take most of the energy dissipation. In intermediate length scales (corresponding to the inertial subrange), both the TKE contribution and dissipation are much smaller but nonzero.

Fig. 1.5 Turbulent energy spectrum as a function of wavenumber

RANS is the most widely adopted simulation method in the industry. Instantaneous balance equations are averaged to get the equations for mean quantities. In this method, only the large vortices are directly solved, and a wide range of turbulence scales are presented by turbulence models [29]. This method is time-saving and computationally cheaper, and it can be applied to complex structures. The selection of models is vitally important as it determines the accuracy of simulations. However, flow field
unsteadiness, which is particularly important when vortical structures develop, cannot be obtained by RANS due to the time averaging of local flows [30].

Compared with RANS, LES is more accurate in time, and the turbulent structures are better presented. Filters are used to divide the turbulent flow into large scales and small scales. The large eddies which cover the majority of the energy spectrum in Fig. 1.5 are solved directly, while the small eddies where most dissipation takes place are required to be modelled [11]. This method has been extensively applied in the study of the dynamics of non-reacting flows in complex structures, and it is promising for turbulent flame simulation. An accurate description of flame properties, such as turbulent transaction and instabilities, can be achieved through LES. Recently, many studies have applied LES to combustion in complex geometries [31-33]. However, the length scale requirements of LES are more stringent than RANS, and the chemical requirements are extremely challenging [25]. Because LES is more expensive than RANS in terms of computational time and resources, a significant number of LES studies are still limited to laboratory scale.

In DNS, governing equations are solved directly, without any turbulence models. As shown in Fig. 1.5, the whole range of turbulence scales, from Kolmogorov length scale to integral length scale, are resolved directly. DNS can adopt detailed chemical kinetics and transport properties in simulation. Consequently, it can be used as a powerful tool that improves the understanding in turbulent combustion’s fundamentals. Besides, DNS can aid the development and validation of turbulent combustion models for engineering application. However, due to the high-resolution requirement and associated high computational demand, the computation domain is usually very simple,
and the size is limited to a few millimetres in three-dimensional simulations or a few centimetres in two-dimensional simulations [34].

Although direct numerical simulation is computationally expensive and limited to simple configurations, this method can help to gain physical insight into the complex processes governing turbulent combustion [35]. A lot of useful information, such as fuel consumption rate, heat release rate, flame stretch and curvature can be provided by DNS, while it is difficult or impossible for the experiment to capture these properties. Moreover, DNS results can also be used to verify combustion models and LES codes. Therefore, DNS study of flames at high turbulence intensities and under elevated pressures will not only improve our understanding of combustion fundamentals, but also develop and validate combustion models. From 2000, three-dimensional DNS of turbulent reacting flows with multi-step chemical mechanisms and transport properties has been realised due to the development of massively parallel computing systems.

Several previous DNS studies were concentrated in the turbulence-flame interaction over a wide range of Karlovitz numbers [36-42]. However, because of the numerical difficulties and high computational cost, limited effort has been dedicated to the analysis of pressure effects at high Karlovitz number.

1.4 Objectives of this research

The purpose of this research is to investigate the turbulence-flame interactions of lean premixed turbulent flames over a wide range of Karlovitz number under elevated pressures using three-dimensional direct numerical simulation. This study will enhance our understanding of the physical and chemical phenomenon of combustion under
conditions relevant to practical combustion devices. The specific objectives are as follows:

a. Investigate the flame structures and chemical pathways of turbulent flames transiting from the thin reaction zone to the distributed reaction zone at elevated pressures. Identify the effects of turbulence intensity on turbulence-flame interactions under elevated pressures.

b. Examine the flame structures and chemical pathways of high-Ka lean premixed flames at the atmospheric and elevated pressures. Identify the effects of pressure on turbulence-flame interactions for flames in the distributed reaction zone.

c. Explore the response of heat release to equivalence ratio variations in lean premixed flames under conditions relevant to modern stationary gas turbines. Identify the reaction zone structures and species distributions at various lean conditions.

d. Gain insight into the effects of integral length scale on turbulence-flame interactions in turbulent flames. Identify flame structures and chemical pathways in turbulent flames with varying ratios of integral length scale to flame thickness under different pressures.

1.5 Thesis outline

This thesis contains eight chapters and the structure is demonstrated in Fig. 1.6. Chapter 1 provides the background and objectives of this research. Chapter 2 summarises the existing research results on premixed combustions at high Karlovitz numbers and under elevated pressures. Chapter 3 provides the mathematical background and numerical methodology of this research. The direct numerical
Simulation results and discussion are presented in Chapters 4, 5 and 6, where Chapter 4 investigates the effects of Karlovitz number, Chapter 5 explores the effects of pressure, Chapter 6 focuses on the effects of equivalence ratios and Chapter 7 discusses the effects of integral length scale. Finally, Chapter 8 summarises this research and provides a future research plan.

Fig. 1.6 A flowchart of the structure of the thesis
Chapter 2

Literature Review

This thesis focuses on lean premixed combustion at high Karlovitz numbers under elevated pressures. In this section, basic concepts of premixed combustion and recent research results about the effects of Karlovitz number and pressure are reviewed.

2.1 Premixed combustion

For diesel engines and traditional gas turbines, the typical type of flame is diffusion flame, where the fuel and oxidiser are not mixed before ignition. However, the pollutant emissions from non-premixed combustion of fossil fuel have drawn great concerns from the public and government. To reduce pollutant emission, alternative technologies for energy conversion have attracted global attention over the past decades. A promising combustion technology is the lean premixed burning in stationary gas turbines, which supplies a good strategy for the control of pollutant emissions [43]. Moreover, this technology can also improve the efficiency of combustion devices. In premixed combustion, one important parameter is the equivalence ratio ($\phi$), which is defined as [10]:

$$\phi = \frac{(F/A)}{(F/A)_{\text{stoic}}}$$

(2.1)
where \( F \) and \( A \) are respectively the mass of fuel and air. \((F / A)_{\text{stoic}}\) is known as stoichiometric air-fuel ratio, which means the quantity of oxidiser equals to the amount of oxygen needed for complete reaction of the fuel and there is no \( \text{O}_2 \) left in the products. When \( \phi \) is lower than unity, it means the mixture is fuel lean and the combustion is known as fuel-lean combustion, or lean combustion.

As mentioned previously, lean premixed combustion is a promising technology which is characterised by low pollutant emissions and high combustion efficiency. As a result, it has been widely employed in industrial combustion devices, including international combustion engines, gas turbines and furnaces [14]. For example, in aero gas turbines, the NOx emission can be reduced by 30-40% when lean premixed combustion technology is adopted [44]. However, the problems associated with lean premixed combustion, e.g. flashback, autoignition and combustion instability, increase operating cost, decrease operating efficiency and limit its application. Thus, it is necessary to improve our understanding of lean premixed combustion and address these fundamental problems in experimental and numerical studies.

2.1.1 Laminar premixed combustion

Laminar premixed flames are the simplest premixed flame, but the understanding of laminar premixed flames is a prerequisite to analyse turbulent premixed flames. Laminar flame speed and thickness are two important parameters to describe a laminar premixed flame. Generally, the laminar flame speed \( S_L \) equals to the movement speed of flame front relevant to the unburnt gas. It is mentioned by Lai [45] that the unstretched adiabatic laminar flame speed is determined by temperature, pressure and species mass fractions. The laminar flame thickness is usually given by:
2.1 Premixed combustion

\[ \delta_T = \frac{(T_b - T_u)}{|\nabla T|_{\text{max}}} \]  

(2.2)

where \( T_b \) and \( T_u \) are respectively the temperatures of burnt and unburnt mixtures. \( |\nabla T|_{\text{max}} \) is the maximum temperature gradient across the flame.

Figure 2.1 presents the structure of a laminar lean premixed H\(_2\)/air flame, which was simulated using the PENCIL code [46] with a multi-step chemical mechanism [47]. The unburnt gas temperature is 298 K, the equivalence ratio is 0.4 and pressure is 1 atm.

![Schematic illustration of the structure of a laminar premixed H\(_2\)/air flame.](image)

Generally, a laminar premixed flame has two zones: the preheat zone and reaction zone [48]. In the preheat zone, the mixture temperature is increased due to heat conduction, while the temperature is still insufficient to maintain reactions. The reaction zone includes several layers, and the first important layer is the inner layer where most of the chain-breaking reactions happen. As can be seen from Fig. 2.1, in the inner layer (grey region), the temperature does not reach the peak point, whereas the mass fraction of radicals (e.g. H) increases dramatically. It is noted that this layer is very thin, whereas the temperature is high enough to maintain chemical reactions. Moreover, the inner layer thickness can be affected by pressure. According to the estimation by Peters [49],
the ratio $l_\delta / \delta_L$ decreases from 0.1 to 0.03 with increasing pressure from 1 atm to 30 atm, where $l_\delta$ is the inner layer thickness and $\delta_L$ is the thermal thickness of laminar flame. The H₂ consumption rate (FCR) and heat release rate (HRR) are also given in Fig. 2.1. As a multi-step mechanism is used in this simulation, the HRR layer does not overlap with the distribution of FCR, and the FCR layer is thinner than the HRR layer. The reactions with chemical radicals can further release heat in the downstream oxidation layer.

The first experimental study of laminar premixed flames can be traced back to mid-19th century when Bunsen designed a burner to measure flame burning velocity [50]. Subsequently, several experimental configurations and methods were proposed to study premixed combustion properties. Especially, there were numerous studies focusing on the flame speed of different fuels under various equivalence ratios, temperatures and pressures [50-57]. It was shown in Refs. [57-59] that the laminar speed of H₂/O₂ increases with increasing equivalence ratio until a slightly fuel-rich condition, then the flame speed decreases with increasing equivalence ratio. Due to the difficulties in performing high-pressure experiments, our understanding of pressure effects on laminar flames is limited to slightly elevated pressures. Zhu et al. [51] studied CH₄/O₂ flame speed under varying pressures from 0.25 atm to 2 atm. It was found that the flame speed decreases with increasing pressure for both fuel-lean and fuel-rich flames. Dayma et al. [60] showed that pressure effects on laminar H₂/O₂ flame speed depend on the equivalence ratio. The flame speed decreases with increasing pressure from 1 to 2 bar at $\varphi = 0.5$, whereas it increases with increasing pressure from 0.3 to 1.7 bar at $\varphi = 1.7$. 

Moreover, accurate measurement of laminar flames plays an important role in the development and validation of kinetic models [61-63]. However, reliable experimental data at high pressures are insufficient, which limits the model optimisation for high-pressure applications [50]. It is also noted that various aspects of laminar flames, e.g. radical concentrations, are difficult or impossible to be studied through experiments. Thus, numerical analysis is adopted to address these issues. Currently, there are several commercial CFD software to simulate laminar flames [64]. Various numerical studies focusing on laminar flame speed, stretch effects and other flame properties have been reported with the aim to validate chemical mechanisms and enhance our understanding of laminar premixed flames [63, 65-71]. Hassan et al. [66] studied laminar premixed CH₄/air flames both experimentally and numerically. The numerical predictions of laminar flame speeds agree very well with experimental results with equivalence ratios of 0.6-1.3 and at pressures of 0.5-3 atm. Natarajan et al. [72] investigated the effects of strain rate on laminar H₂/CO/O₂ flames at pressures up to 5 atm. It was found that the flame speed increases linearly with strain rate at both atmospheric and elevated pressures.

2.1.2 Turbulent premixed combustion

In most combustion devices, the flows are turbulent, which are characterised by random fluctuations of flow properties. Generally, a mean quantity and a fluctuating quantity are used to characterise the properties of a turbulent flow. For instance, the mean velocity in a point is defined by time-averaging of flow velocities over a time interval [10]:

\[
\bar{u}(x) = \frac{1}{\Delta t} \int_{t_i}^{t_f} u(x, t) dt
\]  (2.3)
where \( u(x,t) \) is the instantaneous velocity, and the time interval \( \Delta t = t_2 - t_1 \) should be sufficiently long.

Then, the velocity fluctuation \( u'(x,t) \) is given as [10]:

\[
    u'(x,t) = u(x,t) - \overline{u}(x)
\]  

(2.4)

The above manner is referred to as the Reynolds decomposition, which is also used to present other quantities, e.g. density, pressure and temperature, in turbulent flames.

![Schematic illustration of velocity versus time at a fixed point in a turbulent flow](image)

Fig. 2.2 Schematic illustration of velocity versus time at a fixed point in a turbulent flow, from Ref. [10, 73].

To show the natural characteristic of isotropic turbulence, a root-mean-square (RMS) turbulent velocity is introduced to present velocity pulsations [73]:

\[
    u' = \sqrt{\frac{u_i'^2 + u_j'^2 + u_k'^2}{3}}
\]  

(2.5)

where \( u_i, u_j \) and \( u_k \) are the velocity components in x, y and z directions. When analysing a turbulent flame, the RMS velocity is usually adopted to characterise the
2.1 Premixed combustion

velocity pulsations. Figure 2.2 illustrates the relationship between mean velocity, velocity fluctuation and RMS velocity.

Another technique used to describe a property in turbulent flames is the probability density function (PDF). For example, the PDF of velocity is defined by:

\[ \int_{u_{\min}}^{u_{\max}} P(u) \, du = 1 \]  

(2.6)

where the velocity varies between \( u_{\min} \) and \( u_{\max} \).

Based on PDF, cumulative distribution function (CDF) is also introduced in turbulent flame analysis:

\[ f(u) = \int_{u_{\min}}^{u} P(u') \, du' \]  

(2.7)

In turbulent combustion, any flame properties could be characterised by PDF and CDF over space or over a sufficiently long time interval.

Turbulent combustion is a complicated process that contains numerous physical and chemical phenomenon. In turbulent flames, the turbulent flow could stretch and wrinkle the flame front, which increases flame surface area and modify flame speed. Meanwhile, the chemical reactions lead to the increase in temperature, which increases local viscosity and affects the dissipation of turbulent eddies. As turbulent flames occur in most practical combustion devices, understanding of turbulent combustion fundamentals is crucial in engineering design. In the past decades, experimental and numerical approaches are combined to investigate turbulent premixed flames [74-78]. It is reviewed by Lipatnikov et al. [79] that, the turbulent flame speed under moderate-
intensity turbulence increases with increasing RMS velocity, laminar flame speed and pressure, while the turbulent flame thickness is mainly controlled by turbulent diffusion. Hamlington and Driscoll et al. [80, 81] demonstrated that the preheat zone is substantially broadened with increasing turbulence intensity, whereas the reaction zone is only slightly broadened. Driscoll et al. [74] experimentally studied the vorticity evolution in premixed flames under different turbulence intensities. In their experiment, the viscous diffusion has a weak effect on large vortices, while the effect is significant if the vortices size is reduced. Moreover, they studied the effects of flame on turbulence. The flame-generated vorticity suppresses flame wrinkles and helps to stabilise the propagating flame. Besides, several experimental methods have been proposed by his group to study the dynamic of turbulent premixed flames [82-84].

However, experimental studies of turbulent flames are limited by operational requirements and available diagnostics [85]. With the development of supercomputer, direct numerical simulation of turbulent flames becomes feasible. Poinsot et al. [86-88] studied the stretching and quenching of premixed turbulent flames using direct numerical simulations. A new approach was proposed to define flame-vortex interactions. Chen et al. [89-95] have done numerous fundamental studies of turbulent flames using direct numerical simulation, and several problems, e.g. autoignition, curvature effects and strain rate effects, are clarified in both simple and complex turbulent flows. Wang et al. [96, 97] studied turbulent flame speed and flame structures of lean premixed CH₄/air flames under various turbulence intensities. They found that the turbulent flame speed is relative to the turbulent flame front area. With increasing turbulence intensity, the flame front is seriously curved and the flame area is enhanced. Correspondingly, the turbulent flame speed increases. Sankaran et al. [92] studied 3D
Bunsen CH₄/air flames using DNS and found that the average flame thickness agrees qualitatively with experimental results. It was also demonstrated in Ref. [75] that direct numerical simulation of turbulent flames with detailed chemistry could predict reasonable results compared with experiments.

2.2 Regime of turbulent premixed combustion

Firstly, some concepts of turbulence are given prior to the introduction of regime diagram. In turbulent flames, there are several length scales to describe the turbulence. The most frequently used scales are macroscale $L$, integral length scale $l_i$, Taylor length scale $l_λ$ and Kolmogorov length scale $η$ [10]. Generally, the four length scales follow the relationship $L > l_i > l_λ > η$. Kolmogorov length scale is the smallest scale in the turbulence, at which energy dissipation takes place. According to Kolmogorov’s theory of turbulence, the viscous dissipation of turbulent energy only happens in the smallest turbulent eddies, while the large eddies are not directly affected [73]. By using dimensional analysis, the Kolmogorov length scale is given by [11, 73]:

$$\eta = (v^3 / \varepsilon)^{1/4}$$  \hspace{1cm} (2.8)

where $v$ and $\varepsilon$ are the kinematic viscosity and dissipation rate, respectively. For dimensional reasons,

$$\varepsilon \propto \frac{u^3}{l_i}$$  \hspace{1cm} (2.9)

Moreover, in turbulent combustion, two dimensionless numbers, the Reynolds number and Karlovitz number, are frequently cited to characterise turbulent flames. They are expressed as [87]:
\[ \text{Re} = \frac{u' l}{v} \]  

(2.10)

\[ \text{Ka} = \frac{t_F}{t_\eta} = \frac{\delta_L^2}{\eta^2} = \frac{\delta_L}{\eta u_K} = \frac{(\varepsilon / \nu)^{1/2}}{S_L / \delta_L} \]  

(2.11)

where \( t_F \), \( t_\eta \) and \( u_K \) are the chemical time scale, Kolmogorov time scale and Kolmogorov characteristic velocity, respectively.

For scaling purposes, \( v = S_L \cdot \delta_L \) is introduced [10]. Therefore,

\[ \text{Re} = \frac{u' \cdot l}{S_L \cdot \delta_L} \]  

(2.12)

\[ \text{Ka}^2 = \frac{u' \cdot \delta_L}{S_L^2 \cdot l} \]  

(2.13)

Fig. 2.3 Regime diagram for premixed turbulent combustion.

In order to deal with the complexities of turbulent combustion, a conceptual framework is needed to characterise the turbulence-flame interactions. Thus, several regime diagrams defined in terms of non-dimensional parameters have been proposed
2.2 Regime of turbulent premixed combustion

(Borghi, 1985; Williams, 1985; Abdel-Gayed and Bradley, 1989; Peters, 1999; Law, 2006) [48, 98]. In particular, the new regime proposed by Peters [99] is widely used to characterise turbulent flames. Figure 2.3 presents Peters’s regime diagram for premixed turbulent combustion.

The line Re = 1 separates the regime of laminar flames from that of turbulent flames. In most practical combustion devices, the flames are characterised by Re > 1 and $u' > S_L$. According to Eq. 2.11, the smallest turbulent eddies are more significant than the flame thickness in the corrugated reaction zone where $Ka < 1$. Therefore, the turbulent fluctuation has little effect on the reaction zone structures, and the flame remains quasi-steady [48]. The borderline between the thin and corrugated reaction zones is controlled by the Klimov-Williams criteria which is expressed as:

$$Ka = \frac{t_F}{t_\eta} = \frac{\delta^2}{\eta^2} = 1$$  \hspace{1cm} (2.14)

As such, the Kolmogorov length scale is smaller than the laminar flame thickness in the thin reaction zone, which means the smallest turbulent eddies could penetrate into the reaction regions. The upper boundary of the thin reaction zone is given by $Ka = 100$. Here, the inner layer thickness $l_\delta$ as discussed in Fig. 2.1 is reintroduced to define the reaction zone Karlovitz number $Ka_\delta$:

$$Ka_\delta = \frac{l_\delta^2}{\eta^2} = \delta^2Ka$$  \hspace{1cm} (2.15)

where $\delta = l_\delta / S_L = 0.1$ at the atmospheric pressure. The upper limit of the thin reaction zone can also be expressed as $Ka_\delta = 1$. Therefore, the small eddies can perturb the reactive-diffusive flame structures but cannot enter into the inner layer in the thin reaction zone.
When the RMS velocity is further increased, the flame moves into the broken reaction zone where the Kolmogorov eddies are smaller than the thickness of the inner layer. As a result, the small-scale eddies can enter into the inner layer and reduce local temperatures. As most of the chain-breaking reactions happen in this layer, the chemical pathways may be modified by turbulence, and flame extinction may occur.

As mentioned in Section 2.1.2, turbulent premixed flames have been extensively studied via both experimental and numerical approaches. However, most of the studies focus on the flames in the corrugated and thin reaction zones. Especially, it is challenging and expensive to do experiments under high turbulence intensities and high pressures relevant to combustion in practical devices. In practical combustion devices, the turbulent velocity fluctuation is as high as 150 times of the laminar flame speed and the pressure can reach up to 30 bar [26, 27]. To date, only a few research groups have the ability to set up experiments under extreme conditions [98]. Thus, it is necessary to find an alternative method to investigate the fundamental mechanisms of turbulent combustion under strong turbulent conditions and elevated pressures. With the increasingly powerful supercomputer, direct numerical simulation of turbulent reaction flow at high Ka and under high pressures has become a reality. The following sections will explore the existing literature on high Ka flames and high-pressure combustion via direct numerical simulation.

2.3 DNS of turbulent premixed flames at high Karlovitz numbers

In the past ten years, there has been an increasing number of DNS studies focusing on turbulent premixed flames with high Karlovitz numbers. This section highlights the
2.3 DNS of turbulent premixed flames at high Karlovitz numbers

effects of Karlovitz number on flame structures, turbulent flame speed, flame stretching and chemical pathways. Figure 2.4 summarises the existing DNS studies in a turbulent combustion regime diagram.

Fig. 2.4 Combustion regime diagram showing high-Ka flames. The source of DNS data: Aspden et al. [36-38, 100-102], Carlsson et al. [39, 41], Wang et al. [42, 95, 103], Savrad et al. [104-108], Lapointe et al. [40, 109], Bobbitt et al. [110], Fru et al. [111], Hamlington et al. [80], Poludnenko et al. [112] and Nilsson et al. [113].

Aspden et al. [36-38, 100, 101] have explored lean premixed H₂/air and CH₄/air flames over a wide range of Karlovitz number, and the transition to distributed burning was examined using detailed direct numerical simulation. It was pointed out that the small-scale structures are increased, and the burning rate is intensified with increasing Ka. For high Ka flames, the preheat zone is obviously thickened, while the reaction zone remains unaffected. Especially, distributed flame structures were observed in their studies. In the distributed flames, it was demonstrated that the interface between unburnt and burnt gas becomes smooth, and the reaction rate is reduced under high
Lewis number while increased under low Lewis number. Carlsson et al. [39, 41] studied flame structures of a CH₄/air flame and an H₂/air flame in the broken reaction zone. Due to the effects of turbulent transport and differential diffusion, heat releases in low-temperature regions are significantly enhanced. Turbulent eddies were observed protruding deeply into the burnt gas, whereas the reaction zones are still connected. Fru et al. [111] analysed spherical premixed CH₄/air flame structures under different Karlovitz numbers. It was demonstrated that under low and moderate turbulence intensities, the flame wrinkling grows with increasing Ka, while it stops growing after a certain high value of Ka. Correspondingly, bending effect was observed in the profiles of turbulent flame speed. Hamlington et al. [80] explored the turbulence-flame interactions in turbulent H₂/air flames from the thin reaction zone to the broken reaction zone. They also demonstrated that the thickness of reaction zone is insensitive to turbulence intensities.

Nilsson et al. [113] further increased the Karlovitz number to 3350 in lean premixed CH₄/air flames. It was revealed that the strain rate, reaction and diffusion co-determine the local flame thickness in turbulent flames. Nevertheless, the effects of reaction term are significantly reduced under a ultra-high Ka. Wang et al. [95, 103] have explored the turbulence-flame interactions in a laboratory high Ka premixed CH₄/air jet flame using direct numerical simulation. The DNS results showed that the scale of wrinkling structures is significantly reduced, which provides a good prediction of flame curvatures compared with experimental flames. In another study [42], they examined the flame structures, and it was found that flame thinning occurs in regions with high-positive tangential strain rates while flame thickening happens in regions with high curvatures. In the H₂/air flames, Hamlington et al. [80] noted a similar
phenomenon that the turbulent straining decreases the flame thickness with increasing turbulence intensity. It is also revealed that the effects of flow geometry on turbulence-flame interactions are significant [103]. Through the study of H₂/air flames with both low and high Karlovitz numbers, Savard et al. [104] suggested that the Peters’s regime diagram should be modified for fuels with ultra-low Lewis numbers.

The effects of Lewis number have also been investigated in heavy hydrocarbon flames at high Ka. Through the examination of enstrophy transformation, it was suggested that a detailed chemical mechanism is unnecessary and unity Lewis number transport is enough to capture the main phenomenon of turbulence-flame interactions [110]. Nevertheless, Savard et al. [40, 105, 106] later pointed out that local extinction happens in the flame with non-unity Lewis number and the effects of turbulence on flame structures are controlled by effective Lewis number. In high Ka flames, the reaction zone is obviously broadened, while the inner layer thickness relatively remains unaffected. It was also identified that the effects of differential diffusion are reduced with increasing Ka.

Aspden et al. [36, 38] revealed that the effects of Ka on turbulent flame speed are related to the fuel type and equivalence ratios. For CH₄/air flames, the global flame speed increases with Ka due to the increased flame surface area. However, in H₂/air flames, the turbulent flame speed shows a decreasing trend with increasing Ka after a certain Ka value. This phenomenon is known as the bending effect [114]. In the DNS study of CH₄/air planar flames carried out by Nivarti et al. [115], the mechanism of bending was explored. It was revealed that the inhibited growth of flame surface area directly suppresses the turbulent flame speed at a high Ka. With increasing turbulence
intensity, the number of small-scale eddies is dramatically increased, which results in a highly curved flame front with more negative curvatures. Meanwhile, the efficacy of turbulence straining is reduced. The combined effect is that the increase of flame surface area is suppressed in high Ka flames.

In a high Ka jet flame, Wang et al. [116] have explored the effects of curvature on the displacement speed. In local regions with high curvatures, the effects of curvature term on displacement speed are dominant. However, if the PDF of curvatures shows the mean curvature is zero, the net contribution from curvatures could be neglected. Hawkes et al. [117] studied the effects of H$_2$ blending on CH$_4$/air flames at high Karlovitz numbers. The hydrogen-enriched flame is characterised by a higher flame speed and thus the flame stability is increased. They also revealed that CO emission is reduced while NO emission is increased when H$_2$ is added to the CH$_4$/air flame. For heavy hydrocarbon flames, it is reported by Savard et al. [105, 109] that the turbulent flame speed of a non-unity Lewis number flame is significantly smaller than that in a flame with unity Lewis number, and the flame speed increases in the flame with lighter fuel. At high Ka, the effects of differential diffusion on the strain rate and flame curvatures are limited.

Aspden et al. [37] demonstrated a decorrelation between fuel consumption and heat release at high Ka in H$_2$/air flames, which does not exist for low Ka flames. The flame front regions with high-positive curvatures are characterised by relatively high levels of fuel consumption but low levels of heat release. In recent studies [102], they revealed that the decorrelation of fuel consumption and heat release is solely attributed to the diffusion of atomic hydrogen. Carlsson et al. [39] investigated the evolution of chemical time scales for different species, and revealed that the chemical pathways in
a high Ka H₂/air flame are obviously modified while that in a high Ka CH₄/air flame are only slightly affected. However, Dasgupta et al. [118] analysed the heat release contribution and fuel consumption contribution of several elementary reactions in H₂/air flames at different Karlovitz numbers, and they found that the chemical pathways even remain quite similar between the laminar flame and the turbulent flame in the broken reaction zone.

2.4 DNS of turbulent premixed flames under elevated pressures

Due to the practical interest, combustion under elevated pressures has been the research focus for several years. One of the critical subjects is ignition under engine conditions, which is crucial for the safety and efficiency of high-pressure combustion. There has been a growing body of numerical studies on the ignition and subsequent flame propagation using direct numerical simulation. In the 2D DNS of H₂/air ignition carried out by Chen et al. [119], it was demonstrated that the instantaneous displacement speed is inversely proportional to the temperature gradient, and ~40% of front length is in deflagration mode under conditions relevant to HCCI engines. In another study [120], they reported that the combustion duration and ignition timing can be changed by temperature fluctuation and the length scale of temperature is characterised by a positive correlation with flame front thickness. When turbulence is introduced, the flame front thickness changes slightly with decreasing length scale. Bisetti et al. [121] explored the effects of differential diffusion during the ignition of H₂/air under HCCI engine conditions. In the early phase of autoignition, the differential diffusion of H₂ can increase local temperature and heat release in highly-curved regions.
In the late phase of autoignition, differential diffusion increases heat release in regions with positive curvatures, while decreases heat release in regions with negative curvatures. Yoo et al. [122] further analysed the ignition of \( n \)-heptane/air mixture, and they found that both the mean and fluctuation of temperature determine the critical degree of thermal stratification for smooth operation of HCCI engines. They also explored the ignition characteristics of different fuels [123-126]. Bansal et al. [127] then extended to 3D DNS of autoignition in turbulent DME/air mixtures. The ignition process is identified with three stages and each stage corresponds to a distinct chemical pathway. It is also demonstrated that thin flame structures can be generated in DME autoignition despite the intensity of initial turbulence. It should be noted that all the above studies focus on the ignition process under high pressures (~40 atm), which is performed in a constant volume with pressure-rising conditions. However, this kind of configuration is not suitable to isolate the effects of pressure.

In the past ten years, there have been several DNS studies focusing on the effects of pressure on turbulent flames under elevated pressures. Figure 2.5 summarises the existing DNS studies in a turbulent combustion regime diagram. Reddy et al. [128] investigated the turbulence-kernel interactions in 2D lean CH\(_4\)/air flames with an initial pressure of 40 atm. The turbulent length scale and RMS velocity co-determine the deformation degree of flam fronts. Cecere et al. [129] demonstrated 2D CH\(_4\)/H\(_2\)/air slot flames under pressures of 0.1-4 MPa. Significant small-scale flame wrinkling was observed in the high-pressure flame, which corresponds to a high flame surface density. Dinesh et al. [130] studied the effects of pressure on spherical syngas flames via 3D direct numerical simulation. When pressure is elevated, the spherical flame is severely wrinkled, the flame cells are getting smaller and high-curvature regions are increased.
As a result, the scatter distribution of heat release rate is enhanced at elevated pressures. Moreover, the elevated pressure has a stronger effect on the formation of chemical radicals compared with turbulence intensity. In another study [131], they investigated the effects of differential diffusion on syngas spherical flames at 4 bar. Under low turbulence intensity, thermo-diffusive instability promotes the forming of small-scale structures. When the turbulent intensity is increased, differential diffusion effects are overwhelmed by turbulent mixing effects. Later on, the effects of equivalence ratio were explored in spherical H\textsubscript{2}/CO/air flames from fuel-lean to fuel-rich conditions under 4 atm [132]. It was revealed that Darrieus-Landau instability is insensitive to equivalence ratio, while thermo-diffusive effects are enhanced with increasing equivalence ratio. Wang et al. [133] analysed the effects of pressure on H\textsubscript{2}/air flames in the thin reaction zone. The flame front shows more small cells and thin finger structures when pressure is elevated.

Savard et al. [108] introduced a reaction zone Karlovitz number to explore pressure effects in iso-octane flames under conditions relevant to spark-ignition engines. It was indicated that the heat release rate is more sensitive to curvatures under low pressures due to the combined effects of differential diffusion and local reactions, given that the reaction zone Karlovitz number is constant under different pressures. Recently, Wang et al. [96] have carried out a serious of DNS studies of planar CH\textsubscript{4}/air flames under conditions relevant to lean-burn engines. With increasing Karlovitz number, the regions with high curvatures increases, whereas the local surface area is prone to decrease. In another study [134], they revealed that the Karlovitz number has a significant effect on the small-scale wrinkling, while the large-scale wrinkling is almost independent of Ka on the flame front.
For expanding spherical H$_2$/CO/air flames, it is demonstrated that the turbulent flame speed increases with increasing pressure, and the differential diffusion effect can accelerate the propagation of turbulent spherical flames [130, 131]. Cecere et al. [129] investigated CH$_4$/H$_2$/air flames from the atmospheric pressure to 40 atm. The turbulent flame speed increases with increasing pressures, even though the laminar flame speed decreases as pressure increases. Savard et al. [108] compared the effects of turbulent flame surface area and stretch factor on turbulent flame speed. It was revealed that the normalised flame speed is closely related to the normalised surface area, while the effects of stretch factor are negligible. Wang et al. [96, 97] investigated displacement...
speeds of CH₄/air flames at 20 bar over a range of turbulence intensities and length scales. It was demonstrated that the turbulent flame speed (scaled by laminar flame speed) is strongly related to the flame front area (scaled by inflow cross-sectional area), and the efficiency factor remains close to unity. It was also indicated that the scaled displacement speed by flame front curvature is weakly dependent on the Karlovitz number, whereas it increases with increasing turbulent Reynolds number. In another study of Wang et al. [134], they identified the effects of equivalence ratio on turbulent flame speed, and it was revealed that the primary effect is through the modification of laminar flame speed and the Karlovitz number.

Dinesh et al. [132] revealed that the heat release rate increases with increasing curvature in spherical syngas flames under fuel-lean conditions, whereas an opposite trend was observed when the flames were under fuel-rich conditions. In planar H₂/air flames [133], it was demonstrated that the heat release is enhanced in high-positive curvature regions while reduced in high-negative regions when pressure increases from 1 atm to 5 atm. In laminar flames, the heat release contribution from pressure-dependent reaction H + O₂ (+M) = HO₂ (+M) increases with increasing pressure, while it decreases in turbulent flames. In 3D DNS studies of H₂/air flames under pressure rising conditions, Yenerdag et al. [138] reported that the flame thickness decreases and heat release increases with increasing pressure. After the flame impinging the walls, the wall heat flux was observed to be proportional to pressure. Savard et al. [108] reported that the increased pressure has less effects on the mean heat release rate when the reaction zone Karlovitz number is fixed. They also revealed that the global performance of chemical pathways is independent of pressure in turbulent flames.
Chapter 3

Mathematical, Physical and Numerical Models

This chapter presents the governing equations along with numerical models for direct numerical simulation of premixed flames. Additionally, the direct numerical simulation methodology is also provided.

3.1 Governing Equations

In reaction flows, the governing equations are referred to as the balance equations of mass, momentum, species and energy. In this study, these equations are expressed in the form of advective derivatives with regards to compressible hydrodynamic flows [4, 46, 139, 140]. The continuity equation governs the conservation of mass for the mixture:

$$\frac{D \ln \rho}{Dt} = -\nabla \cdot U$$

(3.1)

where $\rho$ is the mixture density, $\frac{D}{Dt} = \frac{\partial}{\partial t} + U \cdot \nabla$ is the advective derivative and $U$ is the velocity.

The momentum equation is written as:
\[ \frac{DU}{Dt} = \frac{1}{\rho} (\nabla p + F_{vs}) + f \]  

(3.2)

where \( p \) is the pressure and \( f \) is body force in the vertical direction (e.g. buoyancy).

\( F_{vs} \) is the viscous force, which is given by [46, 141]:

\[ F_{vs} = \nabla \cdot (2\rho v S) \]  

(3.3)

where \( S \) is the trace-less rate of strain tensor:

\[ S_{ij} = \frac{1}{2} \left( \frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i} \right) - \frac{1}{3} \delta_{ij} \nabla \cdot U \]  

(3.4)

where \( \delta_{ij} \) is the Kronecker delta, such that \( \delta_{ij} = 1 \) if \( i = j \) and \( \delta_{ij} = 0 \) if \( i \neq j \).

The kinematic viscosity \( v \) is relevant to the dynamic viscosity \( \mu \) [4]:

\[ v = \frac{\mu}{\rho} \]  

(3.5)

In the mixture, the dynamic viscosity of a single component is expressed as [4, 46, 140]:

\[ \mu_k = \frac{5}{16} \frac{\sqrt{\pi M_k k_B T}}{\sigma_k^2 \Omega_k^{(2,2)\rho}} \]  

(3.6)

where \( M_k \) is the molecular weight of species \( k \), \( k_B \) is the Boltzmann constant, \( T \) is the temperature, \( \sigma_k \) is the Lennard-Jones collision diameter and \( \Omega_k^{(2,2)\rho} \) is the collision integral. Transport properties for polar and nonpolar gases are supplied with the chemical mechanism to calculate these parameters.

Then, the mixture viscosity is given by [46, 140]:
where $N$ is the total species number in the mixture, $X_k$ is the mole fraction of species $k$ and $\Phi_{kj}$ is given by:

$$\Phi_{kj} = \frac{1}{\sqrt{8}} \left( 1 + \frac{M_k}{M_j} \right)^{-1/2} \left( 1 + \frac{\mu_k}{\mu_j} \right)^{1/2} \left( \frac{M_j}{M_k} \right)^{1/4}$$  

From Eqs. 3.5 - 3.8, it can be seen that the dynamic viscosity is independent of pressure, while the kinematic viscosity decreases with increasing pressure due to the change of mixture density.

The balance equation for species $k$ is [46, 139]:

$$\frac{d}{dt} \rho Y_k = -\nabla \cdot J_k + \dot{\omega}_k$$  

where $Y_k$, $J_k$ and $\dot{\omega}_k$ are respectively mass fraction, diffusion flux and reaction rate of species $k$. In section 3.3.2, the detailed expression of $\dot{\omega}_k$ is given. $Y_k$ obeys the following condition to achieve mass conservation:

$$\sum_{k=1}^{N} Y_k = 1$$  

The diffusion flux is expressed as:

$$J_k = \rho Y_k V_k$$  

where $V_k$ is the species diffusion velocity. In this study, the mixture averaged approximation is adopted for species diffusion, where Soret effect and body force are neglected, such that $V_k$ is expressed as:
\[ V_k = -\frac{D_k}{X_k} \left( \nabla X_k + \left( X_k - Y_k \right) \frac{1}{p} \nabla p \right) \]  

(3.12)

where \( D_k \) is the diffusion coefficient for species \( k \), which is given by:

\[ D_k = \frac{1-Y_k}{\sum_{j \neq k} X_j / D_{jk}} \]  

(3.13)

where \( X_j \) is the mole fraction for species \( j \) and \( D_{jk} \) is the binary diffusion coefficient which is expressed as [46, 140, 142]:

\[ D_{jk} = \frac{3}{16} \sqrt{\frac{2\pi k_B T^3}{M_{jk}}} \]  

(3.14)

where \( M_{jk} \) is the reduced molecular weight, \( \sigma_{jk} \) is the reduced Lennard-Jones collision diameter, \( \Omega^{(1,1)}_{jk} \) is the collision integral dependent on the reduced temperature for the \((j,k)\) species pair and \( P \) is the pressure. It is obvious that the diffusion coefficient shows an inverse correlation with pressure.

The conservation equation of energy is given by:

\[ \left( c_p - \frac{R}{\bar{M}} \right) \frac{D \ln T}{Dr} = \sum_k \frac{DY_k}{Dr} \left( \frac{R}{M_k} - \frac{h_k}{T} \right) - \frac{R}{\bar{M}} \nabla \cdot U + \frac{2\nu S^2}{T} - \frac{\nabla \cdot q}{\rho T} \]  

(3.15)

where \( c_p \) is the heat capacity, \( R \) is the universal gas constant, \( \bar{M} \) is the molecular weight of the mixture and \( h_k \) is the enthalpy. The heat flux \( q \) is given by:

\[ q = \sum_k h_k J_k - \lambda \nabla T \]  

(3.16)

where \( \lambda \) is the thermal conductivity of the mixture and it is independent of pressure.
For an individual species, the thermal conductivity is composed of translational, rotational and vibrational contributions, which is given by [46, 140, 143]:

$$\lambda_k = \frac{\mu_k}{M_k} \left( f_{\text{trans},k} C_{\text{trans},k} + f_{\text{rot},k} C_{\text{rot},k} + f_{\text{vib},k} C_{\text{vib},k} \right)$$

(3.17)

Then, the mixture-averaged thermal conductivity is expressed by a combination averaging formula [46, 140, 144]:

$$\lambda = \frac{1}{2} \left( \sum_{k=1}^{N} X_k \lambda_k + \frac{1}{\sum_{k=1}^{N} X_k / \lambda_k} \right)$$

(3.18)

In addition to the conservation equations, a state equation is needed to close the equations. In this work, ideal gas equation is applied:

$$p = \rho RT \sum_{k=1}^{N} \frac{Y_k}{M_k}$$

(3.19)

3.2 Turbulence generation

In the present simulations, homogeneous isotropic turbulence is generated with a forcing function proposed by Brandenburg [145]:

$$f(x,t) = \Re \left\{ N f_{k(t)} \exp[i k(t) \cdot x + i \phi(t)] \right\}$$

(3.20)

where \(k(t) = (k_x, k_y, k_z)\) is a time-dependent wavevector and \(\phi(t)\) with \(|\phi| < \pi\) is a random phase. The normalisation factor chosen on dimensional grounds is expressed as:

$$N = f_0 c_s \left( \frac{k c_s}{\delta t} \right)^{1/2}$$

(3.21)
where \( f_0 \) is a non-dimensional coefficient, \( k = |k(t)| \) and \( \delta t \) is the length of time step.

### 3.3 Description of chemistry

#### 3.3.1 Reaction mechanism

The present study only focuses on the combustion of lean premixed H\(_2\)/air. There have been several H\(_2\)/O\(_2\) kinetic models, and the selection of kinetic mechanism has a great influence on the accurate prediction of flame characteristics, especially at elevated pressures. Moreover, the number of species in the mechanism directly determines the computational resource consumption. Thus, it is necessary to select a proper H\(_2\)/O\(_2\) chemical mechanism for the current simulations.

The hydrogen oxidation mechanism developed by Li et al. [47] is one of the most widely used H\(_2\)/O\(_2\) kinetic models in combustion simulation. Ströhle and Myhrvold [146] evaluated several detailed H\(_2\)/O\(_2\) reaction mechanisms under gas turbine conditions, and they found that Li’s mechanism showed the best agreement with experimental data in the investigation of both ignition laminar flame speed and delay time under gas turbine conditions. Therefore, Li’s mechanism is adopted in this study.

In this mechanism, there are 21 reversible reactions and 9 species. Table 3.1 presents the 21 fundamental reactions.

#### 3.3.2 Reaction rate

As the fuel is gas hydrogen, gas-phase chemical rate expressions are introduced in this study [10, 25, 147]. The thermochemical data and other coefficients used in the calculation are supplied by the mechanism.
Considering the forward and reversible reactions, an elementary reaction is usually expressed in the form:

\[
\sum_{k=1}^{N} v'_{k} W_{k} \leftrightarrow \sum_{k=1}^{N} v''_{k} W_{k}
\]  

(3.22)
where $N$ is the number of chemical species, $i$ denotes the $i$th reaction and $W_k$ presents species $k$. $\nu^{\prime}_{ki}$ and $\nu^*_{ki}$ are the stoichiometric coefficients of species $k$ on the reactants side and products side of reaction $i$, respectively.

The production rate or consumption rate of species $k$ is calculated by summing up the contributions of all elementary reactions. The basic production rate of species $k$ is expressed as:

$$\dot{\omega}_k = \sum_{i=1}^{M} \left( \nu_{ki}^* - \nu_{ki}^{\prime} \right) \left( K_{fi} \prod_{k=1}^{N} [X_k]^{\nu_{ki}^{\prime}} - K_{ri} \prod_{k=1}^{N} [X_k]^{\nu_{ki}^*} \right)$$  \hspace{1cm} (3.23)

where $M$ is the number of chemical reactions. $K_{fi}$ and $K_{ri}$ are respectively the forward and reverse reaction rates of reaction $i$. The forward reaction rate is generally calculated by the Arrhenius expression:

$$K_{fi} = A_i \beta^i \exp \left( \frac{-E_i}{RT} \right)$$  \hspace{1cm} (3.24)

where $A_i$ is the pre-exponential factor, $\beta_i$ is the temperature exponent, $E_i$ is the activation energy and $R_e$ is the gas constant. It should be noted that the gas constant $R_e$ is different from the universal gas constant $R$ in Eq. 3.15. $R_e$ is only used in conjunction with the activation energy $E_i$ with compatible units.

The reverse rate constant is given based on the forward rate by:

$$K_{ri} = \frac{K_{fi}}{K_{ri}}$$  \hspace{1cm} (3.25)
where $K_{ci}$ is determined by thermodynamic properties:

$$K_{ci} = K_{pi} \left( \frac{P_{atm}}{RT} \right)^{\sum_{j=1}^{N} (\nu'_{ui}-\nu'_{ki})}$$ (3.26)

where $P_{atm}$ is the atmospheric pressure. $K_{pi}$ is the equilibrium constant given by:

$$K_{pi} = \exp \left[ (\nu^*_{ki}-\nu'_{ki}) \sum_{k=1}^{N} \left( \frac{S_k^o}{R} - \frac{H_k^o}{RT} \right) \right]$$ (3.27)

where $S_k^o$ and $H_k^o$ are the entropy and enthalpy of species $k$, which are defined as:

$$\frac{S_k^o}{R} = a_1 \ln T + a_2 T + a_3 \frac{T^2}{2} + a_4 \frac{T^3}{3} + a_5 \frac{T^4}{4} + a_\gamma$$ (3.28)

$$\frac{H_k^o}{RT} = a_1 + a_2 \frac{T}{2} + a_3 \frac{T^2}{3} + a_4 \frac{T^3}{4} + a_5 \frac{T^4}{5} + a_6 \frac{T}{T}$$ (3.29)

where the thermodynamic data $a_1 - a_\gamma$ are given in the chemical mechanism.

In table 3.1, reactions 5-8 are referred as three-body reactions, which is usually in the form:

$$\sum_{k=1}^{N} \nu'_{ki} W_k + M \rightleftharpoons \sum_{k=1}^{N} \nu^*_{ki} W_k + M$$ (3.30)

where $M$ presents the third body, which is required to proceed the reaction.

In three-body reactions, the effect of the third body concentration must be considered in the expression of reaction rate:
\[
\dot{\omega}_k = \sum_{i=1}^{M} (\nu_{ki} - \nu_{ki}^') [M]\left( K_{fi} \prod_{k=1}^{N} [X_k]^{\nu_{ki}'} - K_{ri} \prod_{k=1}^{N} [X_k]^{\nu_{ki}} \right)
\] (3.31)

where the contribution rate \([M]\) is given by summing up the contribution rates of all species:

\[
[M] = \sum_{k=1}^{N} a_{k,i} X_k
\] (3.32)

where \(a_{k,i}\) is the contribution rate of species \(k\) in the mixture. If all species contribute equally as third bodies, then \(a_{k,i} = 1\). Otherwise, the coefficients will be specified in the mechanism data.

In table 3.1, reaction 9 and 16 are referred to as pressure-dependent reactions, which is expressed in the form:

\[
\sum_{k=1}^{N} \nu_{ki}' W_k (+M) \Leftrightarrow \sum_{k=1}^{N} \nu_{ki}'' W_k (+M)
\] (3.33)

In pressure-dependent reactions, the forward rate constant is expressed as:

\[
K_{fi} = K_\infty \left( \frac{Pr}{Pr + 1} \right)^F
\] (3.34)

\[
Pr = \frac{K_0[M]}{K_\infty}
\] (3.35)

where \(K_\infty\) and \(K_0\) are the high-pressure limit and low-pressure limit of Arrhenius reaction rate, which are given by:
In this study, $F$ is determined by the TROE form instead of Lindemann form where $F = 1$. The TROE approach is computationally expensive but it is more realistic and compact enough in analysing large reacting systems [148, 149]. In TROE form, the $F$ is expressed as:

$$
\log F = \frac{\log F_{cent}}{1 + \left(\frac{\log \text{Pr} + c}{n - d (\log \text{Pr} + c)}\right)^2}
$$

(3.38)

where:

$$
F_{cent} = (1 - \alpha) \exp\left(\frac{-T}{T^{**}}\right) + \alpha \exp\left(\frac{-T}{T^{*}}\right) + \exp\left(\frac{-T^{**}}{T}\right)
$$

(3.39)

$$
c = -0.4 - 0.67 \log F_{cent}
$$

(3.40)

$$
n = 0.75 - 1.27 \log F_{cent}
$$

(3.41)

$$
d = 0.14
$$

(3.42)

where $\alpha$, $T^*$, $T^{**}$ and $T^{***}$ are specified as auxiliary input in the mechanism.

3.4 Direct numerical simulation

3.4.1 Flow configuration

Figure 3.1 shows the schematic diagram of the computational domain, which is used for all DNS cases in this thesis. As can be seen from Fig. 3.1, it is used to simulate a three-dimensional (3D) freely propagating turbulent premixed flame. In DNS analysis,
different configurations are adopted to investigate different problems of premixed flames, such as the turbulent flame speed, flame topological structure, turbulence-flame interaction and flame-wall interaction [105, 115, 150-153]. Usually, the computation domain of DNS study is small and simple due to its high demand of computational resource. The current configuration, where a pre-generated planar flame freely propagates and interacts with the turbulence, is widely used in literature [39, 41, 154-156] to study the turbulent flame structures and chemical reactions. Recently, it was used to investigate the turbulence-flame interaction under conditions relevant to commercial combustion devices [97, 134]. Therefore, it is reasonable to adopt this configuration to study the flame structures and heat release rates at high Karlovitz numbers and under elevated pressures.

Fig. 3.1 Schematic diagram of the computational domain. The red area indicates the flame front.

As shown in Fig. 3.1, the domain size is $2L \times L \times L$ in x, y and z directions, respectively. For all simulations, uniform grids with constant $\Delta x$ in all three directions are used. To avoid the effects of boundary on reaction zone, the domain length is usually set 20 times of the integral length scale in this study, which is higher than that reported in Refs. [39, 41, 154, 155]. The detailed information will be given in the section of
3.4 Direct numerical simulation

‘Simulation parameters’ in Chapters 4, 5, 6 and 7. To save computational hours and avoid unphysical effects, an isotropic turbulence is pre-generated using the method described in section 3.2. The turbulence is then superimposed into the computation domain to initialise the velocity field of turbulent flame, and it will decay with time in the simulation. The RMS velocity fluctuation $u'$ and integral length scale $l_z$ of the pre-generated turbulence are used to characterise the turbulent flame. The integral length scale is defined by [11]:

$$l_z(t) = \int_0^\infty F(r,t)dr \quad (3.43)$$

where $r$ is the distance between two points and $f(r,t)$ is expressed as:

$$F(r,t) = 1 - \frac{3}{4} \frac{C}{K} (\varepsilon r)^{2/3} \quad (3.44)$$

where the universal constant $C = 1.5$ and the turbulent kinetic energy is given by:

$$K = \frac{1}{2} \overline{V^2} \overline{V_1} \quad (3.45)$$

For the configuration shown in Fig. 3.1, the ratio of domain length to integral length scale is controlled by $k$ in Eq. 3.20, and $k$ is ranged within 4.5-5 in this study. The vector number $k(t)$ is around 480, and a random vector is selected at each time step. An appropriate value of $f_0$ in Eq. 3.21 is chosen to generate turbulence with a certain intensity. Moreover, one-dimensional (1D) laminar flames are also pre-generated to initialise the chemical structure of the turbulent flames. The 1D flames employ the same chemical mechanism as the turbulent flames, which is introduced in section 3.3.
3.4.2 Boundary conditions

In many DNS studies of turbulent flames at elevated pressure, periodic boundary conditions are typically applied in all directions in order to treat the elevated pressure [126, 128, 157, 158]. However, this application is unphysical. The choice of boundary conditions for high-pressure cases introduces additional difficulties to DNS simulations of turbulent flames. In this research, periodic boundary conditions are applied in the $y$ and $z$ directions, and inlet/outlet boundary conditions are applied in $x$ direction, as shown in Fig. 3.1. To avoid the reflection of acoustic waves and maintain pressure throughout the simulation, the inlet and outlet are specified using the non-reflecting Navier-Stokes Characteristic Boundary Conditions (NSCBC), which was widely used by a number of researchers in the DNS simulation of turbulent flames at high Karlovitz numbers and under elevated pressures [39, 129, 130, 159]. This method was presented by Poinsot and Lele [160] to solve the fully compressible Navier-Stokes equations, and it shows a higher precision and better stability than classical methods. It was later extended by Lodato et al. [161] to a 3D-NSCBC approach which accounts for the convection and pressure gradients in boundary planes. As a result, the flow distortion and boundary reflection are significantly reduced.

3.4.3 The Pencil Code

The DNS code employed in this study is the Pencil Code [162], which is a high-order code designed for compressible hydrodynamic flows where detailed transport properties and chemical mechanisms are adopted. The code is highly modular, and it enables the flexible selection of different physical modules for various problems. Moreover, the code runs efficiently under MPI on parallel computers. This code was
3.4 Direct numerical simulation

extensively used in astrophysics and magneto-hydrodynamics. Its application in premixed turbulent combustion could be found in Refs. [46, 154, 155].

In the code, the governing equations are solved with a six-order explicit finite difference scheme. A fifth-order up-winding scheme is used for the density to avoid unphysical ‘wiggles’ and as a result, the spatial accuracy is fifth-order. For time advancement, a low-storage third-order Runge-Kutta (RK3-2N) scheme is used and a third-order Livermore Solver for Ordinary Differential Equations (LSODE) is used for chemistry equations in this study. For species diffusion, Soret and buoyancy effects are neglected. In lean premixed H\textsubscript{2}/air flames, Soret diffusion could quantitatively change flame speed, species concentration, flame front structures and et al. [163]. When the pressure is elevated, it is demonstrated that the effects of Soret diffusion are enhanced [164]. However, Soret diffusion is a second-order process and it is weaker compared with Fickian diffusion [4]. In high-Ka flames, Soret effect becomes even smaller because turbulent transport dominates over molecular transport. Thus, mixture-averaged transport model is adopted in most of DNS studies of turbulent flames [36, 37, 102, 165-167] and it is revealed that mixture-averaged transport model could retain the qualitative behaviors of lean H\textsubscript{2}/air flames [36, 37, 168]. Moreover, it is demonstrated that the influence of buoyancy is significantly reduced in flames with high turbulence intensity, especially for flames in small scales [133, 169, 170].

Before the turbulent flame simulations, the implementation of Li’s chemical mechanism in Pencil Code was quantitatively validated with Chemkin [171] for a laminar H\textsubscript{2}/air flame with $\varphi = 0.4$ at 1 atm. Figure 3.2 shows the temperature and H\textsubscript{2} profiles obtained through the above two tools. It can be observed that the results from Pencil Code agree well with the results from Chemkin. For more information about the
validation of chemistry implementation in Pencil Code one can be referred to the literature [46].

Fig. 3.2 Temperature and H₂ profiles of 1D premixed flames (φ = 0.4) at 1 atm calculated by Pencil Code and Chemkin.
Chapter 4

Effects of Karlovitz Number in Lean Premixed H₂/air Flames at 5 atm

This chapter presents three-dimensional direct numerical simulations of lean premixed turbulent H₂/air flames in the thin and distributed reaction zones, with the Karlovitz number at 60, 110 and 1,000, and pressure at 5 atm. Flame front structures and chemical pathways are examined in detail to investigate the effects of turbulence on flames at the elevated pressure. Some results are based on simulations presented in Paper 2.

4.1 Simulation details

4.1.1 Simulation parameters

Three three-dimensional (3D) DNS cases were performed to study lean premixed H₂/air flames at pressure $P = 5$ atm in the configuration shown in Fig. 3.1. For case P₅H, the grid resolution is $1152 \times 576 \times 576$ (in x, y and z directions); for cases P₅M and P₅L, the grid resolution is $768 \times 384 \times 384$. Periodic boundary conditions are applied in lateral directions, and Navier-Stokes Characteristics Boundary Conditions (NSCBC) are applied at the inlet/outlet to maintain pressure. In all cases, the temperature of unburned
mixture is 298 K, and the equivalence ratio is $\phi = 0.4$. A one-dimensional laminar flame and three isotropic turbulent boxes with the same integral length scale, $l_t = 0.0315$ cm, but different turbulence intensities were pre-generated at $P = 5$ atm to initialise the computational domain. The domain width, $L$, corresponds to 10 times the integral length scale. Recalling the relations derived by Peters [48], the three cases locate from the thin reaction zone to the distributed reaction zone as shown in the premixed combustion regime diagram in Fig. 4.1.

![Fig. 4.1 Simulation cases located in the turbulent combustion regime diagram.](image)

The laminar flame and turbulent flames were calculated using the same chemical mechanism and transport properties. Table 4.1 lists the simulation parameters. The turbulent Reynolds number and Karlovitz number are defined as:

$$\text{Ka}^2 = \frac{(u' \cdot \delta)}{(S_L \cdot l_t)}$$  \hspace{1cm} (4.1)

$$\text{Re} = \frac{(u' \cdot l_t)}{(S_L \cdot \delta_L)}$$  \hspace{1cm} (4.2)

where $u'$ is the root-mean-square turbulent velocity fluctuation and $S_L$ is the laminar flame speed. The laminar flame thickness $\delta_L$ is defined by:
4.1 Simulation details

\[ \delta_L = \left( T_b - T_u \right) / |\nabla T|_{\text{max}} \]  \hspace{1cm} (4.3)

where \( T_b \) and \( T_u \) are the temperatures of burned and unburned mixtures, respectively.

Correspondingly, the Kolmogorov length scale is defined by:

\[ \eta = l_t \cdot Re^{3/4} \]  \hspace{1cm} (4.4)

Table 4.1 Simulation parameters of turbulent flames with different Ka.

<table>
<thead>
<tr>
<th>Case</th>
<th>P5L</th>
<th>P5M</th>
<th>P5H</th>
</tr>
</thead>
<tbody>
<tr>
<td>( S_L ) (cm/s)</td>
<td>6.53</td>
<td>6.53</td>
<td>6.53</td>
</tr>
<tr>
<td>( \delta_L ) (cm)</td>
<td>0.0315</td>
<td>0.0315</td>
<td>0.0315</td>
</tr>
<tr>
<td>( l_t ) (cm)</td>
<td>0.0315</td>
<td>0.0315</td>
<td>0.0315</td>
</tr>
<tr>
<td>( u'/S_L )</td>
<td>15</td>
<td>23</td>
<td>100</td>
</tr>
<tr>
<td>( \eta ) (( \mu m ))</td>
<td>41</td>
<td>30</td>
<td>10</td>
</tr>
<tr>
<td>( \Delta x/\eta )</td>
<td>0.2</td>
<td>0.27</td>
<td>0.55</td>
</tr>
<tr>
<td>Re</td>
<td>15</td>
<td>23</td>
<td>100</td>
</tr>
<tr>
<td>Ka</td>
<td>60</td>
<td>110</td>
<td>1000</td>
</tr>
</tbody>
</table>

Moreover, the characteristic turbulence time scale is estimated by the large eddy turn-over time:

\[ \tau_t = l_t / u' \]  \hspace{1cm} (4.5)

With the increasing turbulence intensity, the Karlovitz number increases from 60 to 1000 and the corresponding Kolmogorov length scale decreases from 41 \( \mu m \) to 10 \( \mu m \). Due to the finer turbulent scales at high Ka, a higher grid resolution is required,
which is computationally expensive. To save computational cost, the $l_r / \delta_L$ ratio was kept at unity, which is commonly used in other DNS studies [40, 106, 108].

In order to converge the differential equations, the time step $\Delta t$ in this study is controlled by $\Delta t \leq \Delta t_{CFL}$, where $\Delta t_{CFL}$ is the Courant-Friedrichs-Lewy (CFL) condition. Generally, CFL condition is limited by [172, 173]:

$$\frac{|U|}{\Delta x_i} \frac{\Delta t_{CFL}}{\Delta x_i} \leq 1$$ (4.6)

For the cases in this chapter, the time step ranges within $2.5 \times 10^{-9}$ - $5 \times 10^{-9}$ s.

4.1.2 Grid-independence test

In DNS of turbulent flames, a high grid resolution is required to resolve turbulent scales and chemical scales. Lapointe et al. [40] reported that when $\Delta x \leq 2\eta$, the resolution is sufficient to resolve the small-scale turbulence in DNS. For the chemical scales, Lai [45] demonstrated that at least 14 grid points are needed across the flame thickness for DNS with detailed chemistry, and Nikolaou [35] suggested that approximately 20 grid points are needed inside the flame thickness to achieve a stable simulation. In this study, the grid size is controlled smaller than the Kolmogorov scale, and there are 38-58 cells across the flame thickness. Moreover, grid-independence tests are performed for cases P5H and P5M in two-dimensional (2D) simulations. For the grid-independence test cases, grid numbers are doubled in all directions, but all the other properties are kept the same as the baseline cases. Figure 4.2 shows the temporal evolution of integrated heat release rate (HRR) and Fig. 4.3 shows the probability density function (PDF) of HRR which is normalised by the peak HRR of 1D flame.
4.2 Turbulent flame structures

4.2.1 Flame structures

Fig. 4.2 Temporal evolution of integrated heat release rate for cases P5_M, P5_H and the corresponding grid-independence test cases.

Fig. 4.3 Probability density function of normalised heat release rate for cases P5_M, P5_H and the corresponding grid-independence test cases at $t / \tau_l = 4$.

The temporal evolution of integrated HRR in Fig. 4.2 reveals that the refined simulations are in excellent agreement with the baseline cases. The differences shown in Fig. 4.3 are marginal for numerical simulations. Therefore, the results obtained from the baseline cases are grid independent in this study.

4.2 Turbulent flame structures
For flames in the distributed reaction zone, the spatial flame structures are expected to be different from that of flames in the thin reaction zone. Figure 4.4 shows the instantaneous snapshots of temperature, heat release rates and H\textsubscript{2} consumption rates in the regions around reaction zones for the three flames at \( t / \tau_l = 2 \). To describe the local reaction state, a non-dimensional progress variable \( c \) is introduced:

\[
c = \left( \frac{Y_{a,H_2} - Y_{l,H_2}}{Y_{a,H_2} - Y_{b,H_2}} \right)
\]

where \( Y_{a,H_2} \) is the H\textsubscript{2} mass fraction in the fresh mixture, \( Y_{l,H_2} \) is the local H\textsubscript{2} mass fraction and \( Y_{b,H_2} \) is the H\textsubscript{2} mass fraction in the burned gas. The reaction zone is bounded by \( 0.01 < c < 0.99 \).

As expected, the flame fronts are seriously wrinkled by turbulences in all cases, but the structure scales are different under the three Karlovitz numbers. For cases P5\textsubscript{L} and P5\textsubscript{M}, the size of flame cells is large, and the flame edges are blunt. For case P5\textsubscript{H}, there are many small flame structures with sharp edges. It is also noted that the increases of temperature, heat release rate and fuel consumption rate are marginal in regions with \( c < 0.5 \). Hence, we can define the region \( 0.01 < c < 0.5 \) as the preheat zone. The regions bounded by white solid lines are characterised by higher heat release rates than the peak heat release rate of the laminar flame (HRR\textsubscript{1D, peak}) at 5atm, with most of these regions situated in \( 0.5 < c < 0.99 \). It appears that the regions with high heat release rates and fuel consumption rates agree well with regions with high temperature. For cases P5\textsubscript{L} and P5\textsubscript{M}, the regions with peak heat release rate are characterised by peak temperature, whereas it is not apparent in case P5\textsubscript{H}. 
4.2 Turbulent flame structures

Fig. 4.4 Two-dimensional snapshots of regions around reaction zones showing magnitude of temperature, heat release rate and H₂ consumption rate for the cases at different Ka. The solid, dashed and dotted black lines correspond to $c = 0.01$, 0.5 and 0.99, respectively. The solid white line corresponds to the peak HRR of laminar flame.

Furthermore, Fig. 4.5 shows two-dimensional snapshots of mass fractions for H₂, H, OH and HO₂ in the regions around reaction zones of the three flames. The regions with higher HRR than HRR₁D peak are bounded by solid black lines.
Fig. 4.5 Two-dimensional snapshots of regions around reaction zones showing magnitude of species mass fractions. The solid line corresponds to the peak HRR of laminar flame.
The \( \text{H}_2 \) mass fraction is ultra-low before the high heat release regions, which is attributed to a direct effect of differential diffusion. Correspondingly, the concentrations of major radicals are significantly increased in the heat release zone \((\text{HRR} > \text{HRR}_{1D, \text{peak}})\). Combined with the HRR contours in Fig. 4.4, it is noted that the distribution of radical H agrees well with the distribution of heat release rate for all three cases, which indicates that atomic hydrogen can be used as a reliable HRR for turbulent flames at various turbulence intensities under a slightly elevated pressure. In the thin reaction zone, the Kolmogorov scale is ranged within \( l_\delta < \eta < \delta_L \), where \( l_\delta \) is the thickness of the inner layer. Here the smallest turbulence cannot disrupt the inner layer. When \( \eta < l_\delta \) and the corresponding \( \text{Ka} \) is then increased to exceed 100, the small-scale vortices are sufficiently energetic to disrupt the inner reaction zone, and the turbulent flame is supposed to be in the distributed reaction zone \([99]\). In Fig. 4.4, it is noted that, although case \( P5_M \) is located in the distributed reaction zone, it shows analogous structures with those of flame \( P5_L \). As there is no force to maintain the turbulence, the small-scale vortices are rapidly dissipated by viscosity in the upstream of the flame front. In this case, the flame fronts are mainly stretched by large vortices, and the small scales are less important \([87, 88]\). As a result, flame \( P5_M \) shows similar structures as flame \( P5_L \).

To further explain the effects of Karlovitz number on flame structures, the turbulent vortices near the flame front are examined. Figure 4.6 shows the instantaneous snapshots of flame fronts and the vortex structures for the three cases at the elevated pressure \( at t / \tau_i = 2 \). The flame front is identified as the area surrounding the temperature iso-surface which corresponds to the maximum temperature gradient of the laminar
Effects of Karlovitz Number in Lean Premixed H\textsubscript{2}/air Flames at 5 atm

flame, as in [138]. The turbulent vortices are defined by the iso-surfaces of \( Q_- \) criterion [174, 175]:

\[
Q_- = \frac{1}{2} \left( \Omega_y \Omega_y - S_y S_y \right)
\]

\[
= -\frac{1}{2} \frac{\partial u_j}{\partial x_j} \frac{\partial u_i}{\partial x_i} > 0 \tag{4.8}
\]

where \( S_y \) and \( \Omega_y \) are symmetric and antisymmetric parts of the gradient velocity tensor.

In Fig. 4.6, the \( Q_-^* \) denotes the normalised \( Q_- \) values by \( u' \) and \( \eta \) of the unburned mixture. In this plot, the contour level is set with \( Q_-^* = 0.01 \) as in [138].

![Fig. 4.6 Iso-surface of flame fronts coloured by heat release rates and turbulent vortices defined by the \( Q_- \) criterion (\( Q_-^* =0.01 \)) for the flames under different turbulence intensities.](image)

The unit of the HRR is \( \text{erg/cm}^3 \cdot \text{s} \).

In Fig. 4.6, all flame fronts are highly wrinkled, and the convex regions (protruding to unburned mixture) are characterised by higher heat release rates while the concave regions (protruding to burned mixture) are characterised by lower heat release rates. Flame \( P5_L \) and flame \( P5_M \) present quite similar flame structures, despite that the vortex structures of flame \( P5_M \) are slightly smaller. As mentioned above, the small-scale vortices are rapidly dissipated by viscosity, and the turbulence is unable to disrupt the inner reaction zone. As a result, the turbulence-flame interaction is mainly caused by
4.2 Turbulent flame structures

large vortices in flame P5L and flame P5M. When the Ka is further increased to 1000, the size of eddies located at the upstream of the flame front is dramatically reduced, and a significant number of small cusps and pockets are formed on the flame front. The topological structure of the flame front is more complex. The reduced size of cellular structures at high turbulence intensity was also observed by Day et al. [176] in laboratory-scale flames. Moreover, the flame front is no longer continuous, and the isolated flame front fragments with high HRR are protruding to the unburned side.

![Instantaneous streamlines coloured with temperature contours near the reaction zone.](image)

The solid, dashed and dotted lines correspond to $c = 0.01$, $0.5$ and $0.99$, respectively.

For the present case, we assume $l_\delta = 0.05 \delta L$ at 5 atm [10], then the ratio of inner layer thickness to the Kolmogorov length scale is $l_\delta/\eta \approx 5.7$. Although the smallest scale may be dissipated by viscosity, there are small vortices exist with enough energy to affect the reaction zone and even the inner layer. Figure 4.7 shows the instantaneous streamlines across the reaction zone for the flames at different Ka. For cases P5L and P5M, large vortices could be found in the preheat zone. Due to the increased kinematic viscosity at high temperatures, the vortices are substantially suppressed in the reaction zone. As a result, the flow shows laminar characteristics of regular streamlines after the flame front. Nevertheless, a remarkable phenomenon is observed in the case P5H that both large-scale and small-scale vortices could survive after the flame front, even
though the smallest eddies may have been dissipated by the high viscosity due to the increased temperature. Therefore, the small eddies will enhance scalar mixing, and the topological structure of the flame front is more complex at high Ka.

4.2.2 Statistical analysis

In this study, detailed transport properties and chemical kinetics are adopted in the simulation of H2/air flames. Due to the high diffusivity of atomic hydrogen and molecular hydrogen, differential diffusion would significantly influence the flame structures and chemical reactions. To illustrate the effects of differential diffusion on flames at different Karlovitz numbers under elevated pressures, Fig. 4.8 shows the joint probability density function (JPDF) of local equivalence ratio and temperature for both turbulent and laminar flames at the same large eddy turnover time. For H2/air flames, the local equivalence ratio $\phi_L$ is defined as:

$$\phi_L = \frac{Y_H/(2\cdot M_H)}{Y_O/M_O} \quad (4.9)$$

where $Y_H$ and $Y_O$ are the mass fractions of hydrogen and oxygen element respectively, and $M_H$ and $M_O$ represent the molar mass of hydrogen and oxygen element, respectively.

Fig. 4.8 Scatter plots and JPDFs of temperature and local equivalence ratio under different Karlovitz numbers. The solid line denotes the distribution of the laminar flame at 5atm.
4.2 Turbulent flame structures

The solid line corresponds to the distribution of local equivalence ratio in the laminar flame at 5atm. Differential diffusion effects are clearly observed in the 1D flame where the local equivalence ratio is significantly low in the preheat zone, followed by a dramatically increase in the heat release regions. The minimum \( \phi_L \) value is about 0.25 at \( T = 450 \text{ K} \) while the maximum value is about 0.41 under the peak temperature. For the turbulent flames, the scatter distribution covers a broader range from 0.2 to 0.5. Compared with the data of 1D flame, the temperature corresponding to the minimum \( \phi_L \) value in turbulent flames is slightly changed, while the temperature corresponding to the maximum \( \phi_L \) value increases in flame P5L and flame P5M, which indicates that there are some regions with temperatures higher than the adiabatic temperature in both cases. It is noted that the local equivalence ratio distribution for case P5L is quite similar to that of case P5M. However, the flame shows different phenomenon at high Karlovitz number. For the case P5H, the upper threshold value of local equivalence ratio is reduced, while the lower threshold value is increased. Moreover, it is interesting to find that most of the data distribute over the reference line of the laminar flame, toward the direction of lowering the deviation of local equivalence ratio from that of fresh mixture. The effect of turbulence on species diffusion is becoming predominant with increasing turbulence intensity. This phenomenon of unity effective Lewis number is also observed in high Ka H\(_2\)/air flames under the atmospheric pressure [41], and the effects of differential diffusion almost disappear in the distributed H\(_2\)/air flames reported by Aspden et al. [100]. In current studies, even though the hydrogen diffusion effects are reduced at high Ka, the turbulence-flame interaction is still significantly affected by the differential diffusion.
To further examine the effects of Karlovitz numbers on flame structures and heat release, the following statistical analysis will focus on flame fronts which can well present the turbulence-flame interaction in the reaction zone. Figure 4.9 presents the probability density function (PDF) and cumulative distribution function (CDF) of the flame front curvatures which are scaled by the corresponding laminar flame thickness.

Fig. 4.9 (a) PDFs and (b) CDFs of mean curvature of flame fronts under different Karlovitz numbers.

The mean curvature $\kappa$ is defined as:

$$\kappa = -\frac{1}{2} \nabla \cdot \hat{n}$$  \hspace{1cm} (4.10)

where $\hat{n} = -\nabla T / |\nabla T|$ is the normal unit vector of the flame front. Thus, the curvature is characterised by positive values if the surface is protruding to the fresh mixture (convex region), and with negative values if the surface is protruding to the burned gas (concave region).

For all three cases, the PDFs of curvature are roughly normal distributions. As expected, case $P5_L$ and case $P5_M$ give analogous distribution shapes corresponding to
4.2 Turbulent flame structures

the similar flame front structures shown in Fig. 4.6 and Fig. 4.7. When the flame evolves in the high Ka case P5H, the probability of low curvatures is significantly reduced while the high curvature structures are increased. This is due to the increased number of small eddies at high turbulence intensity, which will enhance the scalar mixing and increase curvature. The CDF plot shows that the probability of negative curvature is about 0.45 for all three cases, which indicates that the relative probability of positive curvature to negative curvature is insensitive to turbulence intensity in the current cases at 5atm. It is also noted that the probability of high positive curvatures ($\kappa \delta_x > 3$) is higher than that of high negative curvatures ($\kappa \delta_x < -3$), which implies that there are more convex structures in the flames under different Karlovitz numbers.

![Fig. 4.10 Scatter plots and JPDFs of heat release rate and mean curvatures on the flame fronts under different Karlovitz numbers.](image)

To illustrate the heat release and fuel consumption on the flame front, Fig. 4.10 shows the scatter plots of mean curvature and normalised heat release rate and Fig. 4.11 gives the scatter plots of normalised heat release rate and normalised fuel consumption rate (FCR) where FCR$_{1D, \text{peak}}$ denotes the peak fuel consumption rate of the corresponding laminar flame. It can be seen that the scatter distribution is extensively broadened, and the HRR gradient is becoming smaller under high Ka due to the enhanced turbulent transport, which indicates a sign of transition to distributed burning.
It is known that local extinction may be triggered when the flame is situated in the distributed reaction zone. However, the heat release rate is increasing with increasing Ka and the peak HRR in flame P5H doubles that in flame P5L and P5M in this study. This result is reasonable as the flame is embedded in a fully developed turbulence [36] and small-scale vortices can counteract the influence of flame stretch [87]. Comparing the heat release in convex and concave structures, it is found that the heat release rate is higher in regions with high positive curvatures than that with high negative curvatures, and the heat release is significantly enhanced in high positive curvature regions in the high Ka flame P5H.

Fig. 4.11 Scatter plots of normalised heat release rate and H₂ consumption rate on the flame fronts coloured by mean curvature. The solid line denotes the mean values.

Figure 4.11 presents the distribution of heat release rate against H₂ consumption rate on the flame front. In the high Ka flame P5H, some data points distribute apart from the reference line of mean values, and the distribution covers a large area. This result is consistent with the findings for high Ka flames at 1 atm [37, 102] that the correlation between heat release and fuel consumption is becoming weaker at the high Ka. Moreover, it is interesting to note that points distributing along the lower edge of the scatter distribution region are characterised by high positive curvatures (red dots), whereas the points distributing along the upper edge are characterised by high negative
4.2 Turbulent flame structures

Curvatures (blue dot). In other words, the heat release rate is lower in high positive curvatures (convex) than that in high negative curvature (concave) when their fuel consumption rates are the same. This is due to the intense differential diffusion of chemical radicals near convex boundaries. Nevertheless, the convex structures are protruding to the unburned mixture where H2 concentration and temperature are relatively higher than that in concave regions, such that the overall heat release rate of high positive curvatures is higher as described in Fig. 4.10. It is also noted that the highest HRR does not happen in regions with highest curvatures, which is corresponding to the effect of the Markstein number [108].

Fig. 4.12 Iso-surfaces of $c = 0.01$ (blue), 0.5 (green) and 0.99 (red). $X_{\text{min}}$ and $X_{\text{max}}$ are respectively the minimum and maximum spatial abscissa values for the three iso-surfaces.

As the laminar flame evolves in the turbulent field, the turbulence will interact with the reaction zone and change the structures of flame front. In this study, the thickness of each iso-surface is given by:

$$
\delta_{\text{iso}} = X_{\text{max}} - X_{\text{min}}
$$

(4.11)
where $X_{\text{min}}$ and $X_{\text{max}}$ are respectively the minimum and maximum abscissa values of the corresponding iso-surface.

The thickness of turbulent flame brush is defined as [112]:

$$
\delta_f = X_{3\text{max}} - X_{1\text{min}} \quad (4.12)
$$

where $X_1$ and $X_3$ represent the abscissa values of iso-surfaces at $c = 0.01$ and 0.99 as shown in Fig. 4.12.

Figure 4.13 shows the development of iso-surface areas with progress variable for flame P5L and P5H. A striking observation is that the two flames in different zones of combustion regime diagram show opposite changing trends. The area of iso-surface is increasing with increasing progress variable under low Ka while the area is decreasing with increasing progress variable under high Ka. In the later part of the oxidation layer, there is a rapid decrease of surface area for flame P5H.

![Fig. 4.13 The development of iso-surface area with progress variable for turbulent flames.](image-url)
4.2 Turbulent flame structures

To further examine the flame brushes, the flame brush properties of the laminar flame and turbulent flames at three progress variables are summarised in Table 4.2.

<table>
<thead>
<tr>
<th></th>
<th>Laminar flame</th>
<th>P5&lt;sub&gt;L&lt;/sub&gt;</th>
<th>P5&lt;sub&gt;H&lt;/sub&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>δ&lt;sub&gt;iso&lt;/sub&gt;-0.01 (mm)</td>
<td>-</td>
<td>1.2</td>
<td>1.6</td>
</tr>
<tr>
<td>δ&lt;sub&gt;iso&lt;/sub&gt;-0.5 (mm)</td>
<td>-</td>
<td>1.8</td>
<td>2.0</td>
</tr>
<tr>
<td>δ&lt;sub&gt;iso&lt;/sub&gt;-0.99 (mm)</td>
<td>-</td>
<td>2.3</td>
<td>1.4</td>
</tr>
<tr>
<td>S&lt;sub&gt;iso&lt;/sub&gt;-0.01 (mm&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>L&lt;sup&gt;2&lt;/sup&gt;=9.9</td>
<td>22.8</td>
<td>56.8</td>
</tr>
<tr>
<td>S&lt;sub&gt;iso&lt;/sub&gt;-0.5 (mm&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>L&lt;sup&gt;2&lt;/sup&gt;=9.9</td>
<td>29.6</td>
<td>43.5</td>
</tr>
<tr>
<td>S&lt;sub&gt;iso&lt;/sub&gt;-0.99 (mm&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>L&lt;sup&gt;2&lt;/sup&gt;=9.9</td>
<td>46.6</td>
<td>22.2</td>
</tr>
<tr>
<td>δ&lt;sub&gt;T&lt;/sub&gt; (mm)</td>
<td>0.47</td>
<td>3.2</td>
<td>2.9</td>
</tr>
</tbody>
</table>

*<sup>S</sup><sub>iso</sub> - surface area.

In the preheat zone, the iso-surface area of flame P5<sub>H</sub> is more than double of that in flame P5<sub>L</sub>, whereas the area of P5<sub>H</sub> is dramatically smaller than that of flame P5<sub>L</sub> at c = 0.99. The iso-surfaces in preheat zone, and inner layer are also thicker at high Ka than that under low Ka. However, the thickness of turbulent flame is slightly decreased for the flame evolving at high Ka. This result indicates that the evolution of flame structure is a combined process with both small and large eddies, and the effects of flame on turbulence are significantly different for the low and high Ka flames at the same eddy turnover time. In the current cases, the preheat and oxidation layers are initialised with similar turbulent fields which will then develop under low and high temperatures separately. Under low Ka, the small vortices are rapidly dissipated at the initial stage of flame evolution in high-temperature regions due to the increased viscosity, and then the stretched large vortices are developing with time. Relatively, the development of large eddies in low temperatures regions is lagging due to the low
dissipation rate. As a result, the large-scale stretch is stronger in regions with higher temperature and thus progress variable. In Fig. 4.13, it appears that the iso-surface area is increasing with increasing progress variable. For the flames in the distributed reaction zone, both large and small eddies could survive after the same turnover time as shown in Fig. 4.7. In other words, the small eddies are still playing an essential role in the flame evolution, which will dramatically increase the number of small structures and thus the flame areas. As the viscosity is increasing with temperature and thus progress variable, the small vortices are gradually dissipated. As a result, the iso-surface area is becoming smaller with increasing progress variable.

4.3 Chemical pathways

<table>
<thead>
<tr>
<th>#</th>
<th>Reaction</th>
<th>#</th>
<th>Reaction</th>
</tr>
</thead>
<tbody>
<tr>
<td>R3</td>
<td>$\text{H}_2 + \text{OH} = \text{H}_2\text{O} + \text{H}$</td>
<td>R8</td>
<td>$\text{H} + \text{OH} + \text{M} = \text{H}_2\text{O} + \text{M}$</td>
</tr>
<tr>
<td>R9</td>
<td>$\text{H} + \text{O}_2 (+\text{M}) = \text{HO}_2 (+\text{M})$</td>
<td>R11</td>
<td>$\text{HO}_2 + \text{H} = \text{OH} + \text{OH}$</td>
</tr>
<tr>
<td>R13</td>
<td>$\text{HO}_2 + \text{OH} = \text{H}_2\text{O} + \text{O}_2$</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

In turbulent flames, the interaction of turbulence with flame is affected by turbulence intensities and pressures. As a result, the elementary reactions may be promoted or suppressed when the turbulence intensity and pressure are increased. However, it is unclear if these pathways will be substantively modified, especially at elevated pressures. In the present $\text{H}_2/\text{O}_2$ chemical mechanism, there are 21 reversible reactions, five of which contribute to 87 - 88% of the integrated heat release. Table 4.3 lists the five main reactions. To investigate the effects of Karlovitz number on the modification of chemical pathways, the heat release from these main reactions are examined globally and locally.
4.3 Chemical pathways

4.3.1 Global description

Figure 4.14 shows the heat release contributions of elementary reactions to the integrated heat release for the laminar and turbulent flames at 5atm. The heat release contribution is calculated as:

\[ C_{hr,j} = \int_{V} \int q_i dV / \int_{V} Q dV \]  

(4.13)

where \( q_i \) is the heat release from reaction \( i \), \( Q \) is the accumulated heat release from all elementary reactions and \( V \) is the volume of the computational domain.

Firstly, the results are significantly different between laminar and turbulent flames, which means the chemical pathways of the laminar flame are markedly modified by the turbulence. The heat release contributions from reactions 8 (R8) and 11 (R11) are enhanced while that from reaction 13 (R13) is decreased dramatically. At the elevated pressure, the pressure-dependent reaction 9 (R9) contributes less in the turbulent flames than that in the laminar flame, which agrees with the conclusion in our study of turbulent flames in the thin reaction zone [133]. Secondly, the contribution of reaction 3 (R3) is less sensitive to turbulence intensity in the studied cases. Compared with case P5L, the contribution of reaction 9 decreases slightly in case P5M but increases in case P5H. R11 and R13 show opposite trends with increasing turbulence intensity. It has been reported in Ref. [108] that the global heat release contribution from each elementary reaction is independent of Karlovitz number under a fixed pressure. In comparison, the deviations between the cases at low and high Ka numbers are larger in this study. However, the maximum difference is only 2.8%, which implies the effects
of Karlovitz number on global fractional contribution of elementary reactions are limited even if the flame is in the distributed reaction zone.

![Image](image.png)

Fig. 4.14 Heat release contributions of elementary reactions to the integrated heat release for the laminar and turbulent flames at 5 atm.

### 4.3.2 Local performance

As the global heat release contribution of individual reaction is insensitive to Karlovitz number, it is of great interest to check the local performance of elementary reactions under different Karlovitz numbers. Figure 4.15 compares the relative heat release rates of the main reactions versus progress variable between flames P5\(_L\) and P5\(_H\).

To eliminate the variation of volumetric heat release at different Karlovitz numbers, the local heat release is scaled by the corresponding accumulated heat release \(C_{hr}\) as:

\[
C_{hr,j} = \int_{0}^{1} HR_{r,j} dc
\]  

(4.14)

where \(HR_{r,j}\) is the relative heat release rate of reaction \(i\). As shown in Fig. 4.10, the normalised heat release rate increases with increasing Ka, whereas it does not mean the
relative heat rate is higher under larger $Ka$. The area under a specific line corresponds to the global heat release contribution of this individual reaction.

It is interesting to note that the chemical pathways are locally modified by turbulence. For all reactions, the relative HRR is obviously enhanced in regions with $c < 0.9$, while suppressed in regions with $c > 0.9$, which indicates that the reaction zone is broadened under high $Ka$. For flame $P5t$, significant heat releases occur in range
$0.6 < c < 1$, except for R8 which starts from $c = 0.7$. For flame P5_H, the range is extended to $0.6 < c < 1$ for R8, and $0.4 < c < 1$ for R3, R9, R11 and R13. Compared with the global contribution of R3 in Fig. 4.14, the local modification of chemical pathways in the flame with high Ka is significant. Moreover, it is noted that these relative HRR are promoted or suppressed with different degrees under the same progress variable, indicating another sign of pathway modification.

Fig. 4.16 Local heat release contributions of elementary reactions versus progress variables.
For a more direct comparison, Fig. 4.16 shows the local heat release contributions of elementary reactions versus progress variables in the whole domain. The local heat release contribution is defined as:

\[
C_{\text{local},j} = \iiint_{V_c} \frac{q_i dV}{Q dV}
\]  \hspace{1cm} (4.15)

where \( V_c \) is the volume of regions with a certain value of progress variable.

Corresponding to the extension of regions of relative heat release rate in Fig. 4.15, the lines denoting local heat release contributions are also moving to lower equivalence ratio regions under high Ka. For all reactions, the local contributions are dramatically increased in the preheat zone, even though there are only traces of heat release. In the region \( 0.5 < c < 0.9 \) where the heat release is significant, the local contributions from R3 and R9 increase obviously, whereas the local contributions from R9 and R13 decrease slightly when Ka is increased. For R11, the local contributions are higher in the region \( c < 0.7 \) for flame P5H; then it becomes the same as that of flame P5L. Combined with Fig. 4.15, it is clear that the chemical pathways are locally changed by turbulence for the flame in the distributed zone. The enhanced heat release in the preheat zone indicates a sign of transition to distributed burning.

The modification of chemical pathways is attributed to the change of competition for radical consumptions in elementary reactions. To explore the reasons for this change, the scatter plots and JPDFs of mass fraction of involved species and progress variables are presented in Fig. 4.17 for the two cases P5L and P5H. Overall, the scatter distributions of the four radicals show similar shapes under different Ka, and the differences are not as evident as that of the heat release.
Comparing the mean concentrations for turbulent flames with the concentrations for the laminar flame, it is evident that a large number of radical species are transported to the preheat zone in turbulent flames, and the species distributions are substantively...
changed by turbulence. Hence, the chemical pathways are significantly modified as shown in Fig. 4.14. Comparing the mean concentrations between the two turbulent flames, there are higher mass concentrations for the four radicals at both low and high values of progress variable in the case P5_H. Since there are sufficient H_2 and O_2 in the preheat zone, the elementary reactions are definitely promoted, and the reaction zone is extended to regions with lower progress variables in the high Ka case. Relatively, the reactions in regions with high values of progress variable are suppressed. As mentioned above, differential diffusion is still significant in the studied cases. Under the combined effects of differential diffusion and turbulent transport, the species distributions are spatially changed with different extents, so that the local heat release contributions are modified in the flame with high Ka.

4.4 Flame evolution

In this study, a laminar flame and a turbulent field are pre-generated to initialise a turbulent flame. As a result, the initial conditions of the turbulent flame are nonphysical. When the flame evolves with time, the turbulence is decaying, the effect of initial conditions is reducing, and finally the flame reaches a realistic physical condition. In addition, the grid size and time step in DNS studies are very small, which makes the simulations computationally expensive and limits the length of simulation time. Thus, a proper simulation time should be determined to save computational resources and obtain reliable results. Published DNS cases in similar configurations as the current study are firstly reviewed. Carlsson et al. [39, 41] studied H_2/air flames under a high Ka. They found that the flames develop rapidly in turbulence, and a fully evolved turbulent flame could be obtained within 0.2 < t/τ_f < 0.4. Most of their analysis were
done at $t / \tau_f = 1$ and $t / \tau_f = 2$. Lapointe et al. [12, 40] studied premixed hydrocarbon flames at high Ka, and they found that only a few eddy turnover times are sufficient to remove the initial unphysical conditions. Moreover, it was demonstrated by Rutland et al. [177] that the time is slightly longer than one eddy turnover time for numerical simulation of turbulent premixed flames to achieve a quasi-equilibrium status.

Fig. 4.18 JPDFs of heat release rates and progress variables for flames P5L, P5M and P5H at $t / \tau_f = 1, 2, 3$ and $4$, respectively.
In the present study, the three cases are performed up to 4 turnover times and the results at different times will be compared statistically to find a proper time for the flame evolution. Figure 4.18 shows the JPDFs of heat releases and progress variables at $t / \tau_i = 1, 2, 3$ and 4 for flames P5L, P5M and P5H, respectively.

At the same large eddy turnover time, the overall heat release is stronger, and the distribution is broader at a higher Karlovitz number. For flame P5H, the scatter distribution extends about 0.2 to lower values of progress variable compared with the other two flames. When the flames evolve with time, the heat release rate is increasing, and the scatters become more distributed in all three cases. The progress variable value corresponding to the peak heat release moves from $c = 0.85$ to $c = 0.8$ in cases P5L and P5M, and to $c = 0.75$ in case P5H. It is noted that scatter distributions and JPDFs at time $t / \tau_i = 2$ are statistically similar to that at later times. The distributions change slightly after $t / \tau_i = 3$. Taken together, all the above results indicate that key phenomena under different Ka could be obtained at time $t / \tau_i = 1$ and the quantities are statistically steady after $t / \tau_i = 2$.

![Fig. 4.19 PDFs of mean curvature of flame fronts under different Karlovitz numbers at time $t / \tau_i = 2$ and 4.](image-url)
To further investigate the flame evolution with time, the flame front structure which is identified by PDF of mean curvature is shown in Fig. 4.19 for the three flames at time $t/\tau_f = 2$ and 4. As previous discussion in Fig. 4.9, the probability of high curvatures (both positive and negative) is significantly increased at high $K_a$, and there are more convex regions than concave regions at $t/\tau_f = 2$. These results could also be obtained from the plot at $t/\tau_f = 4$. Moreover, it can be observed that the peaks are inclining to the positive curvature side in all three cases, and the increase of high positive curvature is more significant than that of high negative curvature in case $P5_H$, which indicates that the difference of CDFs between positive curvatures and negative curvatures is enlarged with time advancement. Hence, the effects of high $K_a$ on flame structures are clearly observed at $t/\tau_f = 2$, and these effects become more apparent at the later time.

![Fig. 4.20 Heat release contributions of elementary reactions to the integrated heat release for turbulent flames at $t/\tau_f = 2$ and 4.](image)

Fig. 4.20 Heat release contributions of elementary reactions to the integrated heat release for turbulent flames at $t/\tau_f = 2$ and 4.
Additionally, the global and local performance of chemical pathways are examined to investigate the effects of time on turbulent flame evolution. Figure 4.20 presents the global heat release contributions of elementary reactions to the integrated heat release at $t/\tau_i = 2$ and 4 for flames $P5_L$ and $P5_H$. It is clearly displayed that the global performance of the five reactions has subtle change from 2 turnover times to 4 turnover times. The effects of high $Ka$ on chemical pathways at $t/\tau_i = 2$ are the same as that at $t/\tau_i = 4$. In conclusion, the effects of evolution time on global heat release contributions could be omitted after 2 eddy turnover times.

To further examine the chemical pathways, Fig. 4.21 shows the local performance of the five main reactions for the two cases at $t/\tau_i = 2$ and 4. Reaction 8 is enhanced across the whole domain with increasing time, whereas the other four reactions are slightly suppressed in the oxidation layer. Nevertheless, it is important to note that the same conclusions could be drawn regardless of the different turnover times. In the high $Ka$ flame, chemical pathways are locally modified, with the heat release rate enhanced in regions with low values of progress variable and the heat release rate suppressed in regions with high values of progress variable.

Through the above statistical analysis, it is clear that the effects of $Ka$ on turbulence-flame interactions and the key phenomena of turbulent flames could be obviously observed at two turnover times. With time advancement, the absolute values of different quantities are changing, but the relative trends do no change for flames with different Karlovitz numbers. Therefore, the discussions before section 4.4 are reasonable. In the following chapters, most cases will be performed up to $t/\tau_i = 2$ due to the limitation of computational hours.
Fig. 4.21 Relative heat release rates of elementary reactions versus progress variables for turbulent flames at $t/\tau_i = 2$ and 4.

As mentioned in Section 3.4, the turbulent flame is initialised with an isotropic turbulence field. To check the degree of isotropy within the initial turbulent field, velocity fluctuations in $i$, $j$ and $k$ directions for flames P5L and P5H are listed in Table 4.4.
It is obvious that velocity fluctuations in three directions are almost identical, which agrees with the characteristics of isotropic turbulence. Moreover, the following relations could also be obtained in the isotropic turbulence [178]:

\[
\left( \frac{\partial U_i}{\partial x_i} \right)^2 = \left( \frac{\partial U_j}{\partial x_j} \right)^2 = \left( \frac{\partial U_k}{\partial x_k} \right)^2
\]  
\hspace{1cm} (4.16)

\[
\left( \frac{\partial U_i}{\partial x_j} \right)^2 = \left( \frac{\partial U_i}{\partial x_k} \right)^2 = \left( \frac{\partial U_j}{\partial x_i} \right)^2 = \cdots
\]  
\hspace{1cm} (4.17)

\[
\frac{\partial U_i}{\partial x_j} \cdot \frac{\partial U_j}{\partial x_i} = \frac{\partial U_i}{\partial x_k} \cdot \frac{\partial U_k}{\partial x_i} = \frac{\partial U_j}{\partial x_k} \cdot \frac{\partial U_k}{\partial x_j}
\]  
\hspace{1cm} (4.18)

When temperature files of 1D flames are superimposed into turbulence, the turbulent field is becoming anisotropic. Table 4.5 summarises the velocity fluctuations in three directions for flames P5L and P5H at \( t / \tau_i = 2 \). The velocity fluctuations in cross-flow directions are significantly reduced, especially in case P5L. The difference in case P5H is not as obvious as that in case P5L due to the shorter evolution time at high \( K_a \). It is also noticed that the velocity fluctuations in \( j \) and \( k \) directions are identical, which agrees with the properties of the configuration. To further check the anisotropy of turbulence in turbulent flames, Table 4.5 lists the properties of velocity gradient in
these two flames at $t / \tau_I = 2$. The terms in Eqs. 4.16-4.18 are normalised by the first term of the corresponding equation.

Table 4.5 Velocity fluctuations for flames P5_L and P5_H at $t / \tau_I = 2$

<table>
<thead>
<tr>
<th>Case</th>
<th>$\sqrt{U_i^2}$ (cm/s)</th>
<th>$\sqrt{U_j^2}$ (cm/s)</th>
<th>$\sqrt{U_k^2}$ (cm/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>P5_L</td>
<td>139.5</td>
<td>55.3</td>
<td>56.6</td>
</tr>
<tr>
<td>P5_H</td>
<td>473.0</td>
<td>448.0</td>
<td>448.8</td>
</tr>
</tbody>
</table>

As can be seen from Table 4.6, some ratios are still close to unity, while some ratios are departing from unity, especially for the terms $\left( \frac{\partial U_j}{\partial x_i} \right)^2 / \left( \frac{\partial U_l}{\partial x_i} \right)^2$ and $\left( \frac{\partial U_i}{\partial x_j} \right)^2 / \left( \frac{\partial U_j}{\partial x_j} \right)^2$. It proves that the turbulence is no longer isotropic with the evolution of turbulent flame.

Table 4.6 Properties of turbulence for flames P5_L and P5_H at $t / \tau_I = 2$

<table>
<thead>
<tr>
<th></th>
<th>P5_L</th>
<th>P5_H</th>
<th>P5_L</th>
<th>P5_H</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\left( \frac{\partial U_j}{\partial x_j} \right)^2 / \left( \frac{\partial U_l}{\partial x_i} \right)^2$</td>
<td>0.98</td>
<td>1.00</td>
<td>1.02</td>
<td>0.99</td>
</tr>
<tr>
<td>$\left( \frac{\partial U_k}{\partial x_k} \right)^2 / \left( \frac{\partial U_l}{\partial x_i} \right)^2$</td>
<td>0.98</td>
<td>0.98</td>
<td>1.14</td>
<td>1.22</td>
</tr>
<tr>
<td>$\left( \frac{\partial U_i}{\partial x_k} \right)^2 \frac{\partial U_i}{\partial x_k} / \frac{\partial U_j}{\partial x_j} / \frac{\partial U_j}{\partial x_i}$</td>
<td>1.03</td>
<td>0.97</td>
<td>1.07</td>
<td>1.00</td>
</tr>
<tr>
<td>$\left( \frac{\partial U_j}{\partial x_k} \right)^2 \frac{\partial U_k}{\partial x_j} / \frac{\partial U_j}{\partial x_j} / \frac{\partial U_j}{\partial x_i}$</td>
<td>1.11</td>
<td>0.96</td>
<td>1.12</td>
<td>1.21</td>
</tr>
<tr>
<td>$\left( \frac{\partial U_k}{\partial x_j} \right)^2 / \left( \frac{\partial U_i}{\partial x_j} \right)^2$</td>
<td>1.01</td>
<td>1.00</td>
<td>1.01</td>
<td>1.00</td>
</tr>
</tbody>
</table>
Moreover, it is supposed that the Kolmogorov length scale will decrease with time because there is no force to maintain the turbulence in the studied cases. With the evolution of turbulent flame, small eddies will be quickly consumed, especially in regions with high temperatures. To prove the current DNS is truly solving all the flow scales, local Kolmogorov length scale is checked at \( t / \tau_f = 2 \) and 4 for cases P5L and P5H. Although the relationship of dissipation rate with velocity and length scale is given in Eq. 2.9, the exact definition of dissipation rate is introduced here to calculate the local Kolmogorov length scale [178, 179]:

\[
\varepsilon_0 = 2 \nu S_{ij} S_{ij} = \nu_0 \left( \frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i} \right) \frac{\partial U_j}{\partial x_i} \quad (4.19)
\]

Using Eqs. 2.8, 3.5 and 4.19, the local Kolmogorov length scale \( \eta_{\text{loc}} \) across the simulation domain is determined by:

\[
\eta_{\text{loc}} = \left( \frac{v_0^3}{\varepsilon_0} \right)^{1/4} = \left( \frac{v_0^2}{\left( \frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i} \right) \frac{\partial U_j}{\partial x_i}} \right)^{1/4} \quad (4.20)
\]

Figure 4.22 shows \( \eta_{\text{loc}} \) normalised by corresponding initial Kolmogorov length scales versus progress variable in the two cases at \( t / \tau_f = 2 \) and 4. As viscosity depends inversely on temperature, the local dissipation rate in reaction zone is much higher than that in preheat zone. Thus, local Kolmogorov length scale is increasing with increasing progress variable in the two cases. Within the unburnt regions, the ratio \( \eta_{\text{loc}} / \eta \) ranges from 1-8, then it becomes higher than 10 in reaction zones and finally it reaches 40 in the completely burnt regions. As the turbulence is decaying with time, the distribution
of $\eta_{\text{loc}} / \eta$ is higher at $t / \tau_i = 4$ than that at $t / \tau_i = 2$. It indicates that the grid resolution is becoming better for current DNS cases as the flame evolves in the turbulence. In the initial turbulent filed, $\eta_{\text{loc}} / \eta$ is kept unity across the domain. When temperature profile is superimposed into the turbulence, the ratio $\eta_{\text{loc}} / \eta$ is increasing and the distribution area is enlarging with temperature. It also proves that the turbulence is no longer isotropic with the evolution of turbulent flame.

Fig. 4.22 Scatter plots of normalised local Kolmogorov length scales and progress variables coloured by local temperature.

4.5 Summary
Three DNS cases with the Karlovitz number at 60, 110 and 1000 are performed under pressure 5 atm. The domain size and the integral length scale are kept constant to isolate the effects of $Ka$ from the other controlling parameters. The flame fronts are dramatically stretched by large-scale vortices in all three cases, while high-curvature regions are only induced by small vortices in the flame with a high $Ka$. The relative probability of positive curvature to negative curvature is insensitive to turbulence intensity. It is also observed that the local heat release rate in regions with high-positive curvatures are higher than that in regions with high-negative curvatures when conditioned on the same $H_2$ consumption rate. Moreover, the local chemical pathways can be modified by high-intensity turbulence, whereas the global heat release contributions of elementary reactions are not susceptible to turbulence intensity.
Chapter 5

Effects of Pressure in High-Ka Lean Premixed H₂/air Flames

In industrial combustion devices, the turbulence intensity is up to 150 times of laminar flame speed and pressure could reach more than 30 bar [26, 27]. Under elevated pressure, the flame thickness will be dramatically reduced, which will affect turbulence-flame interactions. However, it is challenging and expensive to conduct experiments at high pressures, thus the understanding of lean premixed turbulent flames at elevated pressures is still insufficient. This chapter presents three-dimensional direct numerical simulations of lean premixed turbulent H₂/air flames under pressures 1, 5 and 20 atm. The flames locate from the thin reaction to the distributed reaction zones, characterised by varied Karlovitz numbers. Flame front structures and chemical pathways are examined in detail to investigate the effects of elevated pressures. Some results are based on simulations presented in Paper 2.

5.1 Simulation details

5.1.1 Simulation parameters
Turbulent flames under different pressures were investigated using three-dimensional direct numerical simulations in the configuration shown in Fig. 3.1. Periodic boundary conditions are used in cross-flow directions, and Navier-Stokes Characteristics Boundary Conditions (NSCBC) are applied at the inlet/outlet to maintain pressure. For all cases, the unburnt gas temperature is 298 K, and the equivalence ratio is 0.4.

Table 5.1 Simulation parameters of turbulent flames under different pressures.

<table>
<thead>
<tr>
<th>Case</th>
<th>P1M</th>
<th>P1H</th>
<th>P5H</th>
<th>P20H</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pressure (atm)</td>
<td>1</td>
<td>1</td>
<td>5</td>
<td>20</td>
</tr>
<tr>
<td>$S_L$ (cm/s)</td>
<td>23</td>
<td>23</td>
<td>6.53</td>
<td>1.66</td>
</tr>
<tr>
<td>$\delta_L$ (cm)</td>
<td>0.06</td>
<td>0.06</td>
<td>0.0315</td>
<td>0.0254</td>
</tr>
<tr>
<td>$u'$ (cm/s)</td>
<td>653</td>
<td>2300</td>
<td>653</td>
<td>166</td>
</tr>
<tr>
<td>$v$ (cm²/s)</td>
<td>0.181</td>
<td>0.181</td>
<td>0.0362</td>
<td>0.00905</td>
</tr>
<tr>
<td>$l_L$ (cm)</td>
<td>0.06</td>
<td>0.06</td>
<td>0.0315</td>
<td>0.0254</td>
</tr>
<tr>
<td>$\delta_L/\Delta x$</td>
<td>38</td>
<td>51</td>
<td>58</td>
<td>51</td>
</tr>
<tr>
<td>Re</td>
<td>28</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>Ka</td>
<td>151</td>
<td>1000</td>
<td>1000</td>
<td>1000</td>
</tr>
</tbody>
</table>

Firstly, Table 5.1 lists the simulation parameters for four cases with increasing pressure from 1 atm to 20 atm. According to the definition of Karlovitz number and Reynolds number in Eqs. 4.1 and 4.2, cases P1H, P5H, P20H are characterised by the same high Re and Ka, which determines that the three cases are located in the distributed reaction zone in Fig. 5.1. To save computational hours and isolate the effects of pressure, the ratio of integral length scale of initial turbulence to laminar flame thickness is kept at unity. The grid resolutions are $1024 \times 512 \times 512$, $1152 \times 576 \times 576$ and
1024×512×512 (in x, y and z directions) for cases P1_H, P5_H, P20_H, respectively. Correspondingly, there are respectively 51, 58 and 51 grids across the thermal thickness of laminar flames. With increasing pressure, the laminar flame speed decreases significantly from 23 to 1.66 cm/s, and the laminar flame thickness decreases from 0.06 to 0.0254 cm. As the kinematic viscosity is inversely proportional to mixture density, it decreases from 0.181 cm²/s at 1 atm to 0.00905 cm²/s at 20 atm. Case P1_M is set with the same velocity fluctuation with case P5_H to investigate the effects of turbulence intensity on flames under different pressures. However, the initial turbulence fields are different for the two cases. For case P1_M, the turbulence is also controlled by \( \frac{l_t}{\delta_L} = 1 \). The grid resolution is 768×384×384 and there are 38 grids across the laminar flame thickness. In the combustion regime diagram, case P1_M is located at the boundary between the thin and distributed reaction zones, and the Karlovitz number is much lower than that of the other three cases.

![Turbulent combustion regime diagram](image)

Fig. 5.1 Turbulent combustion regime diagram.

Secondly, Table 5.2 gives simulation parameters for another set of cases. The unburnt gas temperature is 298 K, and the equivalence ratio is 0.4. Case S20_H is the
same as case P20H in Table 5.1. In cases S1 and S5, the pressures are 1 atm and 5 atm respectively, while the properties of initial turbulence field are identical with that of case S20H. All the properties of laminar flames are the same as that in Table 5.1. As a result, the ratio of $l_t/\delta_L$ decreases from 1 in S20H case to 0.42 in case S1. The Reynolds number and Karlovitz number of case S1 are tiny compared with case S20H. It can be seen from Fig. 5.1, case S5 is located at the boundary between the thin and distributed reaction zones, while case S1 is located close to the laminar flame zone.

Table 5.2 Simulation parameters of turbulent flames with the same initial turbulence but under different pressures.

<table>
<thead>
<tr>
<th>Case</th>
<th>S1</th>
<th>S5</th>
<th>S20H</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pressure</td>
<td>1 atm</td>
<td>5 atm</td>
<td>20 atm</td>
</tr>
<tr>
<td>$S_L$ (cm/s)</td>
<td>23</td>
<td>6.53</td>
<td>1.66</td>
</tr>
<tr>
<td>$\delta_L$ (cm)</td>
<td>0.06</td>
<td>0.0315</td>
<td>0.0254</td>
</tr>
<tr>
<td>$u'$ (cm/s)</td>
<td>166</td>
<td>166</td>
<td>166</td>
</tr>
<tr>
<td>$v$ (cm²/s)</td>
<td>0.181</td>
<td>0.0362</td>
<td>0.00905</td>
</tr>
<tr>
<td>$l_t$ (cm)</td>
<td>0.0254</td>
<td>0.0254</td>
<td>0.0254</td>
</tr>
<tr>
<td>$\delta_l/\Delta x$</td>
<td>30</td>
<td>32</td>
<td>51</td>
</tr>
<tr>
<td>$l_t/\delta_L$</td>
<td>0.42</td>
<td>0.81</td>
<td>1</td>
</tr>
<tr>
<td>Re</td>
<td>3</td>
<td>21</td>
<td>100</td>
</tr>
<tr>
<td>Ka</td>
<td>30</td>
<td>143</td>
<td>1000</td>
</tr>
</tbody>
</table>

5.1.2 Grid-independence test

For the six cases in this chapter, the minimum grid number across laminar flame thickness is 30, which is supposed to be sufficient to resolve flame and turbulence
5.1 Simulation details

structures in DNS with detailed chemistry [35, 45]. To ensure the computational stability, grid-independence tests are also applied in 2D simulations before the 3D cases are performed in this study. Case P5H has been tested in Chapter 4. The Reynolds numbers of cases S1 and S5 are significantly lower than that of case S20H. Thus, only cases P1H and P20H will be tested in this chapter.

Fig. 5.2 Temporal evolution of integrated heat release rate for cases P1H, P20H and the corresponding grid-independence test cases.

Fig. 5.3 Probability density function of normalised heat release rate for cases P1H, P20H and the corresponding grid-independence test cases at $t/\tau_l = 4$.

In the grid-independence test cases, the grid numbers are doubled while all the other properties are kept identical with that of baseline cases. Figure 5.2 shows the
temporal evolution of integrated heat release rate and Fig. 5.3 gives the probability density function (PDF) of heat release rate which is scaled by the peak heat release rate of corresponding 1D flame. For flames P1H and P20H, baseline cases agree well with the grid-independence test cases in the temporal evolution of integrated HRR. A good agreement is also found in the PDFs of HRR in Fig. 5.3 despite the small fluctuations. Therefore, the results obtained from the baseline cases are grid independent in this study.

5.2 Global flame structures

5.2.1 Laminar flames

In the current simulations, pre-generated 1D flames evolve in specified turbulent fields. Figure 5.4 shows the temperature and H₂ mass fraction of pre-generated laminar flames under pressures 1 atm, 5 atm and 20 atm.

![Fig. 5.4 Temperature and hydrogen mass fraction of laminar flames under pressures 1 atm, 5 atm and 20 atm.](image)

To clearly compare theses laminar flames, the abscissa values are scaled by the corresponding laminar flame thickness. The point with maximum temperature gradient
corresponds to the zero value of abscissa (red dash-dot line in Fig. 5.4). It can be seen that the temperature starts to increase from $x = -1.5\delta_L$ and reaches the peak value at around $x = 1.5\delta_L$. When pressure rises from 1 atm to 20 atm, the peak temperature increases from 1418 to 1501 K. However, the hydrogen mass fraction is lower in the reaction zone when pressure is elevated.

![Graph showing heat release rate of laminar flames under pressures 1 atm, 5 atm, and 20 atm.](image)

Fig. 5.5 Heat release rate of laminar flames under pressures 1 atm, 5 atm, and 20 atm.

Figure 5.5 shows the heat release rate of laminar flames under different pressures. As expected, the peak heat release rate is dramatically increased under elevated pressures. However, it should be noted that this is not a linear growth. The peak value of HRR at 5 atm is 7.65 times of that under 1 atm, whereas the peak values of HRR at 20 atm is only 1.46 times of that under 5 atm. The same changing trend is also observed for the flame thickness in Table 5.1. The thermal thickness decreases significantly from 0.06 cm at 1 atm to 0.0315 cm at 5 atm, and then slightly to 0.0254 cm at 20 atm. With increasing pressure, the thickness of heat release zone is decreasing, even though the peak HRR is dramatically high at 20 atm. For example, the heat release zone covers around $1.5\delta_L$ at 1 atm, while the thickness is about $\delta_L$ at 20 atm.
5.2.2 Turbulent flames

Figure 5.6 shows two-dimensional snapshots of temperature, heat release rate, fuel consumption rate and radical H mass fraction near reaction zones of the turbulent flames listed in Table 5.1. The black solid, dashed and dotted lines correspond to $c = 0.01$, $0.5$ and $0.99$, respectively. The solid white line corresponds to the peak HRR of corresponding laminar flame. It is observed that regions with high heat release rate are characterised by large temperature gradient, which is similar to the behaviour of laminar flames. The contour of heat release rate agrees well with that of fuel consumption rate. However, it is found that peak heat release does not happen in regions with peak H$_2$ consumption in flames P$_1$H and P$_1$M with high turbulence intensities. Moreover, it is noted that the heat release rate and H mass fraction display quite similar contour shapes in all cases, which indicate that radical H may be used as an HRR maker. The above phenomenon will be statistically examined in section 5.3.

Comparing the three flames P$_1$H, P$_5$H and P$_{20}$H with high Ka, it is evident that the regions with HRR higher than $\text{HRR}_{1\text{D, peak}}$ are becoming discontinuous and getting narrower when pressure is elevated. At 1 atm, obvious heat release happens in the region $0.5 < c < 0.99$, whereas the area is significantly decreased at elevated pressures. Moreover, the flame front of case P$_1$H is characterised by more small-scale structures compared with case P$_5$H and P$_{20}$H. Comparing the two flames with the same $u'$, it is noted that large structures on the flame front are with the same scales (conditioned on laminar flame thickness), while the high Ka flame is exhibiting more small structures with high curvatures.
Fig. 5.6 2D snapshots of regions around reaction zones showing magnitude of temperature, heat release rate, fuel consumption rate and H mass fraction.
To further examine the effects of pressure on flame front structures, Fig. 5.7 presents instantaneous streamlines across the reaction zones of turbulent flames P1M, P1H, P5H and P20H.

Fig. 5.7 Instantaneous streamlines coloured by temperature contours near the reaction zones of turbulent flames under 1, 5 and 20 atm, respectively. The solid, dashed and dotted lines correspond to $c = 0.01, 0.5$ and 0.99, respectively.

Firstly, the three cases at the same high Ka but different pressures are compared with each other. It is noted that the reaction zones of high Ka flames are seriously wrinkled with vortices existing within the reaction zone. Comparing the two cases P5H and P1H, it is found that the upstream and downstream turbulence show similar structures, even though the fine turbulence structures are different. In both flames, small eddies are sufficiently energetic to survive across the high-temperature reaction zones. However, there are no small vortices after the reaction zone in the case at 20 atm, which
indicates that small eddies could be quickly dissipated at high pressure. Secondly, it is interesting to note that large vortices survive after the reaction zone in case P1M, which is similar to flame P20H. Comparing the two cases P5H and P1M, it is noted that the flame at elevated pressure is highly wrinkled and characterised by more small eddies, even though they have the same absolute turbulence intensity $u'$.

Fig. 5.8 Iso-surface of flame fronts coloured by heat release rate, and turbulent vortices defined by the $Q$ criterion ($Q^* = 0.01$) for the flames under different pressures. The unit of HRR is erg/cm³s.

Figure 5.8 shows the instantaneous snapshots of flame fronts and vortex structures of the four flames above at the same large eddy turnover time. The flame front is identified by the temperature corresponding to the maximum temperature gradient of the laminar flame. The turbulent vortices are defined by Eq. 4.8 and scaled by $u'$ and $\eta$ of the initial turbulence. As can be seen, flame fronts are dramatically distorted and
convex regions are characterised by high heat release rates. At high Ka, the smallest vortical structures are significantly smaller than the apparent structures formed at the distorted flame front. These small eddies are also highly energetic at high turbulence intensity so that they are very likely to enter the flame zone and even the inner reaction layer. Combined with Fig. 5.7, it becomes clear that a significant portion of turbulent eddies survive the reaction zone, even though the smallest scales may have been dissipated by the high viscosity due to increased temperature. However, flame P20H shows different flame structures. Even though the flame edges are quite sharp, there are no small vortex tubes and the vortex scale is larger than that in the other three cases. In the meantime, for flames P1H and P5H, the flame structures and turbulent eddy structures at the two different pressures are not easily distinguishable from these plots. Statistical analysis is needed to further examine the flame structures at different pressures.

To further investigate the effects of turbulence on flame structures, three flames S1, S5 and S20H are performed under different pressures while the initial turbulent fields are identical. Table 5.9 shows the 2D slices of reaction zone, which presents the absolute values of domain width instead of scaled values as those shown in Fig. 5.6. Thus, the reaction zone at low pressure is apparently thicker than that at high pressure. For flames S1 and S5, the left boundary $c = 0.01$ is out of range of the display. As can be seen from Table 5.2, the kinematic viscosity at 1 atm equals to 20 times the kinematic viscosity at 20 atm. Therefore, the eddies will be quickly dissipated at the low pressure, and the interaction between turbulence and flame is enhanced with increasing pressure. As a result, the flame at 20 atm presents a highly wrinkled reaction zone.
Fig. 5.9 2D snapshots of regions around reaction zones showing magnitude of temperature, heat release rate, fuel consumption rate and H mass fraction for cases initialised with the same turbulent filed but different pressures. The black solid, dashed and dotted lines correspond to $c=0.01$, 0.5 and 0.99 respectively. The solid white line corresponds to the peak HRR of laminar flame.
Comparing concave and convex structures, it is found that the convex regions are becoming much thinner and sharper with increasing pressure. This phenomenon is also observed in our previous studies of turbulent flames in the thin reaction zone [133]. Moreover, in case S5 and S20H, the isolines of HRR1D, peak almost overlap with isoline $c = 0.5$ in convex regions which indicate that heat release is stronger in convex regions compared with that in concave regions. Figure 5.10 presents the instantaneous streamlines across the reaction zones for the three flames at different pressures. It is evident that there are more small vortex structures in front of the reaction zone when pressure is elevated. As case S1 is located near the laminar flame zone in the combustion regime diagram, the vortices are too week and will be quickly dissipated. As a result, the flow shows laminar characteristics of regular streamlines after the flame front. For flame S5, the small vortices are dissipated while the large-scale vortex could survive after the reaction zone.

![Instantaneous streamlines coloured by temperature contours near the reaction zones of turbulent flames initialised with an identical turbulent filed under different pressures. The solid, dashed and dotted lines correspond to $c = 0.01$, 0.5 and 0.99, respectively.](image)

5.3 Statistical analysis

This section will statistically investigate the effects of pressure on flame structures and heat release. As case S1 is similar to a laminar flame, it will not be considered in
5.3 Statistical analysis

This section. Figure 5.11 presents the probability density functions (PDFs) and cumulative distribution functions (CDFs) of flame front curvatures for cases under pressures 1 atm, 5 atm and 20 atm. The mean curvatures are scaled by the thickness of the corresponding laminar flames. It is interesting to note that flame structures for cases with the same $K_a$ but different pressures are statistically different, even though flames $P_{1H}$ and $P_{5H}$ show analogous structures in Fig. 5.8. With decreasing pressure, the probability of high curvatures (both positive and negative) is increasing. Especially, the probability of high negative curvatures in flame $P_{1H}$ is significantly increased. In Fig. 5.8, there are more small vortex structures in flame $P_{1M}$ than those in flame $P_{20H}$. However, the probability of high curvatures in flame $P_{20H}$ is higher than that in flame $P_{1M}$. At the initial stage of flame evolution, there are many small vortices in case $P_{20H}$, which will enhance scalar mixing and increase curvature. After a certain time, the small-scale vortices are dissipated, and the flame is mainly stretched by large vortices. However, the effects of small vortices at the initial evolution stage are inherited. Therefore, flame $P_{20H}$ shows a higher probability of high curvatures than flame $P_{1M}$.

Fig. 5.11 PDFs and CDFs of mean curvatures of flame fronts under different pressures.
For the two flames at 1 atm, the cumulative probability of negative curvatures is around 0.51, while the value for the flame at 5 atm is around 0.45. This indicates that the convex regions are enlarged when pressure is elevated, which agrees with the experimental results in [180]. However, it is noted that the increase of convex regions is evident under slightly elevated pressures. As described in Appendix A [133], a similar trend is also observed in turbulent flames in the thin reaction zone as pressure increases from 1 atm to 5 atm. In this study, when pressure is further increased to 20 atm, the probability of negative curvatures even increases slightly to 0.46. Combined with the findings in Chapter 4, it implies that the relative probability of positive curvature to negative curvature is sensitive to pressure, but insensitive to turbulence intensity for high Ka flames.

It should be noted that the following approximation is used in Peters’s definition of Re and Ka [48]:

\[ v = Sc \cdot D = D = S_L \cdot \delta_L \]  

(5.1)

where Sc is the Schmidt number and D is the thermal diffusivity.

The approximation \( Sc = 1 \) is adopted in Eq. 5.1. However, the Schmidt number for the H2/air mixture is around 0.2 instead of 1. Using Peter’s definition of Re and Ka, the ratio of \( \eta / \delta_L \) is identical for the three cases with the same Ka. If the real Sc number is adopted in the definition, the ratio for case P1H is 0.069, while the value for case P5H is 0.086. It means there are relatively more small-scale vortices for the case at the atmospheric pressure. As the high curvatures cusps are induced by small eddies, there should exist larger high curvature areas in the case P1H. It is also mentioned in Ref. [10] that the ratio of inner layer thickness \( l_\delta \) to laminar flame thickness \( \delta_L \) decreases
with increasing pressure. If this phenomenon is taken into account, the boundary between the thin reaction zone and the broken reaction zone shown in Fig. 5.1 should move upward at elevated pressures (as indicated by the red dashed line). It suggests that the relative value of Ka at high pressure is smaller than that at low pressure when the three cases are located at the same point in the regime diagram.

Fig. 5.12 Scatter plots of normalised heat release rate and H\(_2\) consumption rate on the flame fronts coloured by mean curvatures. The solid line denotes the mean values.

Aspden et al. [37, 102] found that the correlation between heat release and H\(_2\) consumption is becoming weaker with increasing Ka for flames at the atmospheric pressure. It is interesting to examine whether the similar phenomenon could be observed at high pressures. Figure 5.12 presents the distribution of normalised heat release rate against normalised fuel (H\(_2\)) consumption rate on the flame front with varying pressures. For case P1\(_H\) at 1atm, the scatters distribute away from the mean.
values and cover a larger area, which agrees with the findings at the atmospheric pressure [37, 102]. With increasing pressure, it is interesting to find that the degree of scattering becomes smaller, which implies that the correlation between heat release and $H_2$ consumption is becoming stronger even though the $Ka$ is high. Moreover, the degree of scattering for case $P1M$ is analogous to that for case $P5H$. It is expected that the decorrelation degree of heat release and $H_2$ consumption is directly related to the turbulence intensity $u'$.

Another striking observation is that points with high curvatures (red points) distribute along lower bound and points with low curvatures (blue points) distribute along upper bound of the distribution region in flames at 1 and 5 atm. It means that the local convex structures with high positive curvatures are characterised by relatively lower HRR, but the concave structures with high negative curvatures are associated with higher HRR, conditioned on the same FCR. However, this phenomenon disappears when pressure is further increased to 20 atm. In flame $P20H$, the heat release rate in high positive curvatures is even higher than that in high negative curvatures. This suggests that the ratio of inner layer thickness to flame thermal thickness reduces with increasing pressure, and it becomes harder for turbulent eddies to disrupt the inner reaction zone. Moreover, the points with the highest curvature values are not located in regions of highest HRR at high turbulence intensity, which corresponds to the effect of the Markstein number [108].

Figure 5.13 shows the CDFs of mean curvatures for flames S5 and S20H. It is found that the probability of negative curvatures in flame $S20H$ is slightly higher than that of flame S5, which agrees with the finding in Fig. 5.11. Even if the two cases are initialised with an identical turbulent field, flame $S20H$ is characterised by more high curvature
regions. The difference between the two cases in high negative curvatures is more obvious than that for flames P5H and P20H.

![CDFs of mean curvatures of flame fronts for cases S5 and S20H.](image)

**Fig. 5.13** CDFs of mean curvatures of flame fronts for cases S5 and S20H.

Figure 5.14 presents the scatter plots of normalised heat release rate and fuel consumption rate on the flame fronts of S5 and S20H. The two cases show a similar degree of correlation between heat release and H\textsubscript{2} consumption. Under 5 atm, the high-positive curvatures are also characterised by lower heat release rate conditioned on the same fuel consumption rate. These findings well support the conclusions from Fig. 5.12.

![Scatter plots of normalised heat release rate and H\textsubscript{2} consumption rate on the flame fronts for cases S5 and S20H.](image)

**Fig. 5.14** Scatter plots of normalised heat release rate and H\textsubscript{2} consumption rate on the flame fronts for cases S5 and S20H.
In Figs. 5.6 and 5.9, the radical H shows a potential to be a heat release rate marker. Therefore, the distribution of H mass fraction against heat release rate is plotted in Fig. 5.15 to examine if H could be used as a reliable marker for turbulent flames at high pressures.

Fig. 5.15 Scatter plots of H mass fraction and heat release rate. The scatters are coloured by local temperatures.

For the three cases at the same Ka, the scatter distribution area increases with decreasing pressure, which implies that the H performs poorly at low pressures. For the
flames at 1 atm, there are even two distribution branches. The temperature is increasing in the clockwise direction for all flames. Moreover, the flames show similar distribution area when they are under the same pressure. For example, the central angle of distribution sector in case S5 is almost the same as that of case P5_H. Therefore, radical H could not be directly used as an HRR marker for high Ka flames under various pressures.

Fig. 5.16 Scatter plots of scaled H mass fraction and heat release rate. The scatters are coloured by local temperatures.
The temperature gradient in Fig. 5.15 implies that the local HRR is also determined by local temperature. Therefore, it is feasible to get a fitting formula which could predict the HRR using both H mass fraction and temperature. As the temperature uniformly changes in the sector regions, a scaled H mass fraction is introduced:

\[ Y_H^* = \frac{Y_H}{T^n} \]  

where \( n = 1, 2, 3, \ldots, N \).

When \( n = 3.6 \), the HRR distribution against scaled H mass fraction is shown in Fig. 5.16. A striking observation is that the scaled H mass fraction shows a linear spatial correlation with HRR in the studied cases, especially for flames at elevated pressures. Therefore, H could be used as a perfect HRR marker for flames with varying pressures when it is scaled by temperature. However, it is noted that the slope is insensitive to turbulence intensity, but increases with increasing pressure. Figure 5.17 displays a fitted curve for pressure and HRR/\( Y_H^* \).

![Graph showing pressure vs. HRR/\( Y_H^* \).](image)

Fig. 5.17 A fitted curve for pressure and HRR/\( Y_H^* \).

In order to provide a universal formula to predict HRR using H mass fraction, three non-dimensional parameters are introduced:
\[ HRR^* = \frac{HRR}{HRR_{\text{max,1D}}} \]  
\[ T^* = \frac{T}{T_0} \]  
\[ P^* = \frac{P}{P_{\text{atm}}} \]

where \( HRR_{\text{max,1D}} \) is the peak heat release rate in the corresponding laminar flame, \( T_0 \) is the unburnt gas temperature and \( P_{\text{atm}} \) is the atmospheric pressure.

Taking the effects of pressure into account, the heat release rate is predicted by:

\[ HRR^* = \alpha \frac{Y_{\text{H}_2} P^*}{T^{3.5}} \]

where the coefficient \( \alpha = 3.5 \times 10^6 \) for cases with \( \phi = 0.4 \).

### 5.4 Chemical pathways

The turbulence-flame interaction is affected by both turbulence intensities and pressures. Chapter 4 has discussed the effects of turbulence intensities on chemical pathways at elevated pressure, while the effects of pressure are not clarified. In the multi-step chemical mechanism, there are several pressure-dependent reactions whose reaction rates are determined by pressure as well as temperature. In the \( \text{H}_2/\text{O}_2 \) mechanism by Li et al. [47] as shown in Table 3.1, reaction \( \text{H} + \text{O}_2 (+\text{M}) = \text{HO}_2 (+\text{M}) \) is a pressure-dependent reaction and it contributes to over 30% of total heat releases. To investigate the effects of pressure on the modification of chemical pathways, the fractional heat release from seven main reactions are firstly examined. Figure 5.18 presents the heat release contributions of elementary reactions versus pressures for both laminar and turbulent flames.
Fig. 5.18 Heat release contributions of elementary reactions against pressures for both laminar (solid lines) and turbulent (dashed lines) flames. The left panel corresponds to turbulent flames initialised with an identical turbulent field under different pressures. The right panel corresponds to turbulent flames with the same Ka but different pressures.

Two sets of cases are compared in Fig. 5.18. The first set includes cases S1, S5 and S20h which are initialised with an identical turbulent field under different pressures. The second set includes cases P1h, P5h and P20h which have the same Karlovitz number but different pressures. As the structure of case S1 is similar to a laminar flow, the heat release contributions in this flame are almost identical to that of laminar flame. It is obvious that the laminar flames are susceptible to elevated pressures. When pressure increases from 1 atm to 20 atm, the heat release contribution from the pressure-dependent reaction \( H + O_2 (+M) = HO_2 (+M) \) increase from 38% to 52%, and the contribution from reaction \( HO_2 + OH = H_2O + O_2 \) dramatically increases from 16% to 34%. The contributions from reaction \( HO_2 + H = OH + OH \) and \( H + OH + M = H_2O + M \) decreases 8-10% with increasing pressure. The other three reactions show little fluctuations in global heat release contributions. In general, the turbulent flames present similar changing trends to the laminar flames. Reaction \( H + O_2 (+M) = HO_2 (+M) \) and
5.4 Chemical pathways

HO₂ + OH = H₂O + O₂ are still the dominate heat release reactions in turbulent flames. However, it shows that the chemical pathways are being changed at elevated pressures, and the phenomenon becomes more significant at higher pressures. For the three cases with the same Ka, the shift of reactions in turbulent flames at 1 atm is quite small, with up to 3% difference. When pressure is increased to 5 atm, reaction HO₂ + OH = H₂O + O₂ presents the maximum change of 9% compared with the laminar flame. Reactions HO₂ + H = OH + OH and H + OH + M = H₂O + M are also relatively enhanced in turbulent flames. When pressure is further increased to 20 atm, significant changes are observed in many reactions. The dominant heat release reactions H + O₂ (+M) = HO₂ (+M) and HO₂ + OH = H₂O + O₂ are suppressed by 8% and 5% respectively in heat release contribution. The fractional heat releases from reaction HO₂ + H = OH + OH and H + OH + M = H₂O + M increases 3% and 4%, respectively. Reactions H₂ + OH = H₂O + H, HO₂ + O = O₂ + OH and H + O₂ = O + OH are less sensitive to pressures. Comparing the two sets of cases, a striking similarity is observed for the changing trend of heat release contributions versus pressure. It indicates that the effects of pressure on global chemical modification are more significant than that of turbulence intensity and Karlovitz number.

Furthermore, the local performance of elementary reactions under different pressures is examined. Figure 5.19 shows the relative heat release contributions of elementary reactions versus progress variable for flames S1, S2 and S20H. It is evident that chemical pathways are locally modified by elevated pressures. The left edge of relative heat release is shifting to higher progress variables with increasing pressure. For example, in the dominant heat release reaction R9, obvious heat release starts from \( c = 0.5 \) at 1 atm, which increases to \( c = 0.7 \) at 20 atm. As a result, the heat release region
is shrinking, and the heat release intensity is becoming higher with increasing pressure, which corresponds to the narrow HRR regions in Fig. 5.9.

Fig. 5.19 Relative heat release contributions of individual elementary reactions versus progress variable for flames initialised with an identical turbulent field under different pressures.

To isolate the effects of pressure from Karlovitz number and turbulence scales, Fig. 5.20 presents the relative heat release contributions of elementary reactions versus progress variable for flames P1H, P5H and P20H. The effects of pressure are more evident and chemical pathways are significantly modified in Fig. 5.20. At \( c < 0.7 \), all the selected reactions are significantly promoted in flames P1H and P5H. With decreasing pressure, the overall heat release is moving to regions with lower equivalence ratios. Note that these reaction contribution rates are modified by different degrees at the same \( c \) value, which illustrates the different effects of pressure on different chemical pathways. As mentioned above, the inner layer in the reaction zone is dramatically decreased at high pressure. It is also known that most of the radical
production happens in the inner layer. Therefore, it becomes harder for small eddies to penetrate into the inner layer and transport species to the low-temperature regions when pressure is elevated.

Recalling the effects of pressure on modification of global chemical pathways, there are significant differences in fractional heat release between the laminar and turbulent flames at 20 atm. It is directly related to the production or consumption rates of chemical radicals. The situation of competition between elementary reactions is changed by pressure. Figure 5.21 shows the contributions of H consumption, OH production and HO$_2$ consumption of elementary reactions versus pressures for the two sets of turbulent flames and laminar flames. Similar to the heat release contributions, the changing trend shown by flames S1, S2 and S20$_H$ resembles that of flames P1$_H$, P5$_H$ and P20$_H$. 

Fig. 5.20 Relative heat release contributions of individual elementary reactions versus progress variable for flames with the same Ka but different pressures.
Fig. 5.21 Contributions of H consumption, OH production and HO₂ consumption of elementary reactions versus pressures for both laminar (solid lines) and turbulent (dashed lines) flames. The left column corresponds to turbulent flames with the same Ka but different pressures. The right column corresponds to turbulent flames initialised with an identical turbulent field under different pressures.
With increasing pressure, the consumption/production of radicals are dramatically modified. The maximum change happens for reaction $\text{HO}_2 + \text{H} = \text{OH} + \text{OH}$ in the laminar flames, with 34% decrease in $\text{HO}_2$ consumption, when pressure increases from 1 atm to 20 atm. This reaction also demonstrates a change of 28% in OH production. Reaction $\text{H} + \text{O}_2 (+\text{M}) = \text{HO}_2 (+\text{M})$ is dominant in H consumption, with 19% increase when pressure is elevated to 20 atm. Compared with heat release, the radical consumption/production shows a higher sensitivity with increasing pressures. Moreover, turbulent flames show a lower sensitivity than laminar flames with increasing pressure. As a result, the differences between laminar and turbulent flames are enlarged when pressure is elevated. In other words, chemical pathways are significantly altered by turbulence for flames at elevated pressures.

5.5 Summary

Two sets of DNS cases with varying pressure are analysed. In the first set, the Karlovitz number, the ratio of domain length to integral length scale and the ratio of integral length scale to laminar flame thickness are kept constant, while the pressure is maintained at 1, 5 and 20 atm, respectively. The second set is initialised with an identical turbulent field but under different pressures. With increasing pressure, the flame speed and thickness decreases, and the reaction zone moves to regions with higher values of progress variable. Moreover, the thickness of inner layer conditioned on the laminar flame thickness becomes smaller under elevated pressures, which results in a lower probability of finding high curvatures in the high-pressure flames with a fixed $K_a$. Nevertheless, the change in PDF distribution of curvature is not significant when the high-pressure flame is initialised with an identical turbulence field as the flame at
the atmospheric pressure. In elementary reactions, pressure could significantly modify the chemical pathways of both laminar and turbulent flames, and the effects are more significant than those of the Karlovitz number.
Chapter 6

Effects of Equivalence Ratios in High-Ka Lean Premixed H2/air Flames at High Pressures

In premixed turbulent flames, equivalence ratio oscillation could trigger combustion instability and local extinction, which will affect the normal operation of combustion devices. In modern gas turbines, typical equivalence ratios at base load are in the range of 0.45-0.6 [43]. Therefore, lean premixed turbulent H2/air flames with equivalence ratio 0.4, 0.5 and 0.6, respectively, are investigated via three-dimensional direct numerical simulation in this chapter. The three flames are all located in the distributed reaction zone and the pressure is 20 atm, which are relevant to gas turbine conditions. Reaction zone structures and heat release rate are examined to explore the effects of equivalence ratios. Some results are based on cases presented in Paper 3.

6.1 Simulation details

6.1.1 Simulation parameters
Lean premixed H₂/air flames were studied using three-dimensional direct numerical simulations in the configuration shown in Fig. 3.1. The initial pressure is 20 atm, the temperature of unburnt gas is 298 K, and the equivalence ratios of unburnt mixture are 0.4, 0.5 and 0.6, respectively. For case A, the grid resolution is 1024×512×512 (in x, y and z directions); for cases B and C, the grid resolution is 512×256×256. In the cross-flow directions, periodic boundary conditions are used; at the inlet/outlet of computational domain, Navier-Stokes Characteristics Boundary Conditions (NSCBC) are applied to maintain the high pressure. Three laminar flames with equivalence ratios 0.4, 0.5 and 0.6, respectively, were pre-generated to initialise the chemistry of turbulent flames. Three isotropic turbulent boxes with integral length scales 2.54×10⁻², 8.3×10⁻³ and 3.3×10⁻³ cm, respectively, were pre-generated to initialise the velocity fields of turbulent flames. The domain length corresponds to 20 times of the integral length scales. As shown in Fig. 6.1, all three cases are located in the distributed reaction zone in a turbulent combustion regime diagram [48].

![Turbulent combustion regime diagram](image)

Fig. 6.1 Turbulent combustion regime diagram.
6.1 Simulation details

The parameters for the cases in this chapter are summarised in Table 6.1.

Table 6.1 Simulation parameters of turbulent flames with equivalence ratios 0.4, 0.5 and 0.6.

<table>
<thead>
<tr>
<th>Case</th>
<th>A</th>
<th>B</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pressure (atm)</td>
<td>20</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>$\phi$</td>
<td>0.4</td>
<td>0.5</td>
<td>0.6</td>
</tr>
<tr>
<td>$S_L$ (cm/s)</td>
<td>1.66</td>
<td>6.81</td>
<td>21.16</td>
</tr>
<tr>
<td>$\delta_L$ (cm)</td>
<td>$2.54 \times 10^{-2}$</td>
<td>$8.3 \times 10^{-3}$</td>
<td>$3.3 \times 10^{-3}$</td>
</tr>
<tr>
<td>$u'$ (cm/s)</td>
<td>177</td>
<td>664</td>
<td>1941</td>
</tr>
<tr>
<td>$v$ (cm$^2$/s)</td>
<td>$9.05 \times 10^{-3}$</td>
<td>$9.34 \times 10^{-3}$</td>
<td>$9.62 \times 10^{-3}$</td>
</tr>
<tr>
<td>$l_t$ (cm)</td>
<td>$2.54 \times 10^{-2}$</td>
<td>$8.3 \times 10^{-3}$</td>
<td>$3.3 \times 10^{-3}$</td>
</tr>
<tr>
<td>$\eta$ ((\mu)m)</td>
<td>2.41</td>
<td>0.69</td>
<td>0.25</td>
</tr>
<tr>
<td>$\delta_L$/$\Delta x$</td>
<td>51</td>
<td>26</td>
<td>26</td>
</tr>
<tr>
<td>Re</td>
<td>497</td>
<td>590</td>
<td>666</td>
</tr>
<tr>
<td>Ka</td>
<td>2376</td>
<td>2373</td>
<td>2368</td>
</tr>
</tbody>
</table>

With increasing equivalence ratios, the laminar flame speed increases significantly from 1.66 to 21.16 cm/s, and the laminar flame thickness decreases from $2.54 \times 10^{-2}$ to $3.3 \times 10^{-3}$ cm. As pressure is constant for the three cases, the kinematic viscosity increases slightly with increasing equivalence ratio. As the definition of Re and Ka in Eqs. 4.1 and 4.2 introduce errors in the estimation of smallest flow structures, they will not be used to define turbulence properties in this chapter. The original definitions of Re and Ka [87] are adopted:

$$\text{Re} = \left( \frac{u' \cdot l_t}{\nu} \right)$$ (6.1)
Effects of Equivalence Ratios in High-Ka Lean Premixed H₂/air Flames at High Pressures

\[ \text{Ka} = \frac{t_F}{t_\eta} = \frac{\delta_L/S_L}{\eta/u_K} = \left( \frac{\varepsilon/\nu}{S_L/\delta_L} \right)^{1/2} \]  

(6.2)

where \( t_F \) and \( t_\eta \) are respectively the chemical time scale and Kolmogorov time scale.

\( u_K \) is the Kolmogorov characteristic velocity. The dissipation rate \( \varepsilon \) is given by:

\[ \varepsilon = u^3/l \]  

(6.3)

To isolate the effects of equivalence ratio from that of other parameters, the ratio of integral length scale to laminar flame thickness is kept at \( l_i/\delta_L = 1 \), and the Karlovitz numbers are kept at around 2370. In flame C, the grid size is dramatically smaller than that in flame A, and the velocity fluctuation is significantly higher. Correspondingly, the time step is very small. Limited by CFL conditions [172, 173], the timestep ranges from \( 1 \times 10^{-10} - 2.5 \times 10^{-9} \) s in this chapter.

6.1.2 Grid-independence test

For the three cases studied in this chapter, there are at least 26 grids across the flame thickness, which is higher than the recommended resolutions in Refs. [35, 45]. Moreover, grid-independence tests are also performed in two-dimensional (2D) simulations. As case A was tested in Chapter 5, only cases B and C will be examined in this chapter. For the grid-independence test cases, grid numbers are doubled in all directions, but all the other properties are kept the same as the baseline cases. Figure 6.2 shows the temporal evolution of integrated heat release rate (HRR) and Fig. 6.3 shows the probability density function (PDF) of HRR which is normalised by the peak HRR of corresponding 1D flames.
Fig. 6.2 Temporal evolution of integrated heat release rate for cases B, C and the corresponding grid-independence test cases.

As can be seen from Fig. 6.2, the integrated heat release increases with time, and there are good agreements in the temporal evolution of integrated heat release between the baseline case and refined case. In Fig. 6.3, there are small fluctuations for the PDFs of heat release rate. However, the differences between the baseline case and refined case are marginal for numerical simulations. Therefore, the results obtained from the baseline cases are grid independent in this study.

Fig. 6.3 Probability density function of normalised heat release rate for cases B, C and the corresponding grid-independence test cases at $t / \tau_l = 4$. 
6.2 Overview of laminar and turbulent flames

As the turbulent flames are initialised with laminar flames, we first discuss the flame characteristics of laminar flames before we explore the turbulent flames. Figure 6.4 shows the temperature and heat release rate (HRR) profiles of laminar flames at equivalence ratio 0.4, 0.5 and 0.6, where the abscissa values are scaled by the corresponding laminar flame thickness. For better comparison, the heat release rates of flame A and B are respectively enlarged to 10 and 5 times of their original values. It is evident that the HRR is significantly increased when the equivalence ratio increases. Table 6.2 lists the key parameters of the reaction zones.

Table 6.2 Characteristic parameters of reaction zones of laminar and turbulent flames

<table>
<thead>
<tr>
<th>Case</th>
<th>HRR_{\text{max,1D}} (\text{erg/cm}^3\cdot\text{s})</th>
<th>Mean HRR_{\text{max,3D}} (\text{erg/cm}^3\cdot\text{s})</th>
<th>\delta_f_{\text{1D}}</th>
<th>\delta_f_{\text{3D}}</th>
<th>T_1 (K)</th>
<th>T_2 (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>7.90e10</td>
<td>3.69e11</td>
<td>0.54 \delta_L</td>
<td>1.25 \delta_L</td>
<td>1188</td>
<td>1403</td>
</tr>
<tr>
<td>B</td>
<td>7.87e11</td>
<td>1.95e12</td>
<td>0.65 \delta_L</td>
<td>1.63 \delta_L</td>
<td>1238</td>
<td>1478</td>
</tr>
<tr>
<td>C</td>
<td>4.68e12</td>
<td>6.73e12</td>
<td>\delta_L</td>
<td>1.86 \delta_L</td>
<td>1196</td>
<td>1543</td>
</tr>
</tbody>
</table>

\(\delta_f\) - reaction layer thickness

\(T_1\) - temperature corresponds to maximum temperature gradient of laminar flames

\(T_2\) - temperature corresponds to peak heat release rate of laminar flames

As expected, the burnt gas temperature increases from 1530 K to 1836 K and the HRR is dramatically enhanced when the equivalence ratio increases from 0.4 to 0.6. Correspondingly, the scaled temperature gradient in Fig. 6.4 is larger under higher equivalence ratio to ensure the same scaled thermal thickness \(\delta_L\). However, it should be
noted that the reaction zone is thinner than the thermal thickness in the studied cases. Hereby, the reaction zone is defined as regions with heat release rate higher than 10% threshold of corresponding laminar flames. When the equivalence ratio increases, the reaction zone is obviously broadened. As shown in Table 6.2, the reaction layer thickness of laminar flames increases from 0.54 \( \delta_l \) to \( \delta_l \) when the equivalence ratio increases from 0.4 to 0.6. Moreover, two characteristic temperatures of laminar flames are presented. The temperature corresponding to maximum temperature gradient fluctuates around 1200 K; the temperature corresponding to peak heat release rate increases with increasing equivalence ratio, from 1403 K to 1543 K.

![Fig. 6.4 Structures of lean premixed laminar flames at different equivalence ratios. The red dashed line denotes the abscissa value which corresponds to the maximum temperature gradient.](image)

Next, the reaction zone characteristics of turbulent flames with \( \varphi = 0.4, 0.5 \) and 0.6 are discussed. Figure 6.5 shows the instantaneous snapshots of temperature, heat release rates, \( \text{H}_2 \) consumption rates and mass fraction of radical H in the regions around reaction zones for the three flames. The slices are taken from \( z = 0.5L \) of the computational domain.
Fig. 6.5 Two-dimensional snapshots of regions around reaction zones showing magnitude of temperature, heat release rates, \( \text{H}_2 \) consumption rates and \( \text{H} \) mass fraction for the cases with equivalence ratios 0.4, 0.5 and 0.6. The solid, dashed and dotted black lines correspond to \( c = 0.01, 0.5 \) and 0.99, respectively.
It is clear that the flame fronts are significantly wrinkled and stretched by turbulences in all cases, and the isolines indicate that the three flames are characterised by similar small and large structures. The majority of heat release happens in region \( 0.5 < c < 0.99 \), even though the heat release rate increases with increasing equivalence ratio. It is also noted that the heat release rate, fuel consumption rate and mass fraction of radical H show quite similar contour shapes in all three cases, which means there is a strong degree of correlation between the three quantities. When the equivalence ratio increases from 0.4 to 0.5, the thickness of high-HRR regions is obviously increased, and the gradient is correspondingly decreased. In flame C, the high-HRR regions almost fill up the zone \( 0.5 < c < 0.99 \), whereas there are only narrow strips in flame A.

![Fig. 6.6 Reaction zone structures of lean premixed turbulent flames at different equivalence ratios.](image)

To further examine the reaction layers, Fig. 6.6 presents 3D structures of reaction layers, Fig. 6.7 shows the mean heat release rate versus progress variable (defined by hydrogen concentration in Eq. 4.7 and Table 6.2 gives the key parameters of turbulent flames with equivalence ratios 0.4, 0.5 and 0.6, respectively. The average reaction layer thickness of turbulent flame is defined as:

\[
l_f = \frac{V_{rec}}{S_{yz}}
\]

(6.4)
where $V_{rec}$ is the volume of reaction zone and $S_{yz}$ is the cross-sectional area of the computational domain.

![Graph](image)

Fig. 6.7 Mean heat release rate versus progress variable for turbulent flames at different equivalence ratios.

Since the three cases are characterised by the same Ka, the flame fronts show analogous structures in Fig. 6.6. Similar to the laminar flames, the heat release of turbulent flames is enhanced, and the reaction layer is relatively broadened under high equivalence ratio. Correspondingly, the reaction zone covers a wider range of temperature at high $\phi$. These differences under various equivalence ratios are attributed to the differential diffusion effect in premixed combustion [181]. In Fig. 6.7, obvious heat release is observed from $c = 0.4$ for case $\phi = 0.6$, while it happens after $c = 0.7$ for case $\phi = 0.4$. It is also noted that the reaction layers of turbulent flames are thicker than the corresponding thermal thicknesses, and the thickness increases with increasing equivalence ratio. Although the turbulent flames exhibit similar changing trends to laminar flames with varying equivalence ratios, the extent of enhancement is reduced at high equivalence ratio. For example, the peak mean heat release rate for the turbulent
flame with $\phi = 0.4$ is 3.7 times higher than that of the corresponding laminar flame, while the value is only 0.4 times for the turbulent flame with $\phi = 0.6$. The reaction layer thickness almost doubles in the laminar flames when the equivalence ratio increases from 0.4 to 0.6. For the turbulent flames, the thickness only increases from $1.25 \delta_L$ to $1.86 \delta_L$. One possible explanation is that the enhanced heat release of turbulent H$_2$/air flame is a combined result of turbulent transport and species diffusion, however, the turbulent transport is overtaking differential diffusion as the main effect when the flame transits to the distributed reaction mode.

### 6.3 Characteristics of turbulent reaction zones

![Two-dimensional slices of the reaction zone coloured by local equivalence ratio. The black dashed lines correspond to temperature gradients, the solid black lines bound the $\phi_L$ plateaus and the solid white lines bound the reaction zone.](image)

In this section, we discuss the local and statistical values regarding the heat release in turbulent reaction zones. Figure 6.8 shows the zoomed-in contours of local equivalence ratio ($\phi_L$) in the flame brush, where the reaction layers ($\text{HRR} > 0.1\text{HRR}_{\text{max},1D}$) are bounded by the solid white lines and the $\phi_L$ plateaus ($\phi_L > 1.1 \phi$) are bounded by the solid black lines. The local equivalence ratio is defined in Eq. 4.9.
The contours of local equivalence ratio indicate the presence of differential diffusion in the studied cases. It is obviously noted that $\phi_L$ is significantly lower than the fresh gas $\phi$ in the upstream near the reaction layer and all the $\phi_L$ plateaus of the three cases locate inside the reaction layer. Hereby, we define the left boundary of the reaction zone as ‘$\phi_L$ cliff’, the convex region towards the unburned side is characterised by positive curvature and the concave region is characterised by negative curvature. It is interesting to find that the $\phi_L$ trenches and plateaus are situated on both sides of the cliff, which is much like the natural topography. The temperature gradient near $\phi_L$ cliff is observed to be high at positive curvatures, and low at negative curvatures. However, the difference in temperature gradient between positive and negative curvatures is reduced with increasing $\phi$. Furthermore, the trench ‘depth’ and plateau ‘height’ are reduced when the equivalence ratio of fresh gas is increased. This is primarily due to the decrease of the Lewis number with increasing $\phi$, which promotes a diffusive-thermally stable flame [182].

Since the $\phi_L$ cliff area is also characterised by a high gradient of temperature in Fig. 6.8, it is necessary to shed more light on the correlation of temperature, $\phi_L$ and heat release rate in the reaction layer. Figure 6.9 shows the scattered $\phi_L$ versus temperature in the reaction zone, where the scatters are coloured by local heat release rate. The adiabatic temperatures of the corresponding laminar flames and equivalence ratios of fresh mixtures are also indicated in the plots. Overall, the scatter distribution shows a positive correlation between $\phi_L$ and temperature for the three cases. Nevertheless, the broader scatter distribution of case $\phi = 0.6$ indicates that the correlation rate is reduced when $\phi$ is increased. It is interesting to note that there are many hot spots above the adiabatic temperature under leaner conditions. For example, for case $\phi = 0.4$, the
adiabatic laminar flame temperature is 1530 K, while the peak temperature of the turbulent flame exceeds 1800 K. A similar result was also reported by Aspden et al. [36] for ultra-lean H₂/air flames. On the other hand, the temperature of the whole reaction zone is below the adiabatic temperature for case $\phi = 0.6$. As mentioned before, lean-premixed combustion has the potential to reduce NOₓ emission due to low peak combustion temperature. However, temperature in the hot regions under ultra-lean conditions (case $\phi = 0.4$) would exceed the adiabatic temperature to a large extent, which will contribute to the formation of NOₓ. This result is instructive and meaningful for industrial application of lean-premixed combustion.

![Fig. 6.9 Scatter plots of local temperature and local equivalence ratio in the reaction zone.](image)

The scatters are coloured by local heat release rate. The horizontal dashed lines denote the equivalence ratio in the unburned gas, and the vertical dashed lines denote the peak temperatures of the corresponding laminar flames.

The presence of hot spots is a consequence of extended $\phi_L$ distribution above unburnt mixture $\phi$ under leaner condition. As indicated in Fig. 6.9, the $\phi_L$ extends to about 0.15 higher than the unburned mixture $\phi$ for case $\phi = 0.4$, while the value is only around 0.05 for case $\phi = 0.6$. As a result, the local heat release rate is relatively enhanced in regions with high $\phi_L$. Furthermore, the heat release rate shows a positive correlation with both temperature and $\phi_L$. Peak heat release happens in regions with high temperature and high $\phi_L$. However, it is observed that the HRR gradient is decreasing.
across temperature with increasing $\phi$. For example, the tail of case $\phi = 0.6$ in Fig. 6.9 is coloured by blue and green (low and moderate HRR), while the tail of case $\phi = 0.4$ is only coloured by blue (low HRR). This observation agrees with the results shown in Fig. 6.8. Moreover, the scatter distribution area is larger especially in high-temperature regions for case $\phi = 0.6$, which agrees well with the broadened heat release region in Fig. 6.5.

To shed more light on the effects of flame structures on the heat release rate, two temperature iso-surfaces in the reaction zone are selected, and the PDFs for scaled curvature and normalised HRR are shown in Fig. 6.10. The two temperatures, $T_1$ and $T_2$ as indicated in Table 6.2, respectively, correspond to maximum temperature gradient and peak heat release rate in laminar flames. The mean curvature is defined by Eq. 4.10. In Fig. 6.10, the mean curvature is scaled by the corresponding laminar flame thickness, and the value is positive for the convex regions towards the unburned side. For iso-surfaces of $T_1$, the scatters show a normal distribution for case $\phi = 0.4$ and 0.5, although the HRR of positive curvatures is relatively higher than that of negative curvatures. Nevertheless, the behaviour of the heat release rate distribution for case $\phi = 0.6$ differs from the other two cases. The lowest HRR occurring in positive curvature regions is lower than that in negative curvature regions; the overall HRR in positive curvature regions is higher than that in negative curvature regions. It should be noted that the PDF gradient is decreasing with increasing equivalence ratio, which means the enhancement (weakening) of reaction rate in convex (concave) regions is reduced as the equivalence ratio increases. This finding is consistent with the results of Luo et al. [181] for H2/air flames under lean and stoichiometric conditions at the atmospheric pressure.
6.3 Characteristics of turbulent reaction zones

Fig. 6.10 Scatter plots of curvature and normalised heat release rate on the iso-surfaces corresponding temperature $T_1$ (left column) and $T_2$ (right column) as indicated in Table 6.2. The curvature is scaled by the corresponding laminar flame thickness.

On iso-surfaces of $T_2$, the aforementioned changing trends still exist and even become more obvious. The scatters are distributing toward the positive side. The HRR in convex regions is significantly enhanced even for case $\phi = 0.6$. Moreover, the peak HRR happens in low curvature regions, which is due to the effects of the Markstein number [108]. Comparing the ordinate values of the two set plots, it is found that the
gap is decreasing with increasing equivalence ratio. For example, the ordinate threshold increases from 15 to 40 as the temperature increases from $T_1$ to $T_2$ at $\phi = 0.4$, while the threshold reaches the same value 5 at $\phi = 0.6$. It indicates that the heat release gradient is specially reduced under high equivalence ratio. Overall, the response of HRR to local curvature is changing with equivalence ratio, and either set of the iso-surfaces can present the trend of change.

![PDFs of mean curvatures of flame fronts for cases under different equivalence ratios. The mean curvatures are scaled by corresponding laminar flame thicknesses.](image)

In Fig. 6.8, the isoline of $T_1$ is very close to the left boundary of the reaction layer. Thus, flame front structures are statistically identified by the $T_1$ iso-surface for the three cases. Figure 6.11 presents the PDFs of flame front curvatures. The mean curvatures are scaled by the thickness of corresponding laminar flames. The statistical result shows that the flame front structures are quite similar in the three flames, despite the peak value for flame $\phi = 0.5$ is a little bit smaller. The main reason is that the three flames with different equivalence ratios have the same Karlovitz number which determines the same chemical time scale conditioned on the Kolmogorov time scale. As a result, the wrinkling and stretching of flame front by turbulence are at the same level. However,
different flame structures are observed for flames with the same Ka but different pressures as shown in Chapter 5. It is noted that the kinematic viscosity changes significantly under various pressures, whereas it increases slightly with increasing equivalence ratios in this chapter. The dissipation rate of turbulent eddies is directly related to the viscosity magnitude. Therefore, the similar values of kinematic viscosities may be the main reason for the analogous flames structures in this study. The effect of kinematic viscosity on turbulence evolutions is an interesting topic for future research.

Fig. 6.12 PDFs of local equivalence ratio gradients for cases under different equivalence ratios. The mean curvatures are scaled by corresponding laminar flame thicknesses.

As aforementioned, the positive-curvature regions are characterised by high temperature and $\phi_L$ gradients, and the gradient reduces with increasing equivalence ratio. To statistically analyse the structures, Figure 6.12 shows the PDFs of local equivalence gradients, where the gradients are referred to as the absolute values of $\nabla \phi_L$ and scaled by the corresponding laminar flame thickness. With increasing equivalence ratio, the probabilities of ultra-high and ultra-low $\phi_L$ gradients decrease. For example, in the ranges $0 < \nabla \phi_L \cdot \delta_L < 1$ and $2 < \nabla \phi_L \cdot \delta_L < 3$, the probability is higher for the flame
with lower equivalence ratio, while it is lower in the range \( 1 < \nabla \phi_L \cdot \delta_L < 2 \). In other words, the difference of gradients in convex and concave regions is reducing with increasing equivalence ratios. This results in the reduced trench ‘depth’ and plateau ‘height’ for case \( \phi = 0.6 \) in Fig. 6.8.

Fig. 6.13 Scatter plots of curvatures and equivalence ratio gradients on the iso-surface corresponding to temperature \( T_1 \). The scatters are coloured by heat release rates.

It is interesting to observe ‘natural topography’ in \( \phi_L \) contours in Fig. 6.8, where the convex structures look like cliffs and the concave structures resemble offshore pools. As a result, the \( \phi_L \) gradients are different in the two kinds of structures. To further examine the spatial structures, Fig. 6.13 presents the scatter plots of curvatures and \( \phi_L \) gradients on the iso-surface corresponding to temperature \( T_1 \). The scatters are coloured by heat release rates. For all three cases, the \( \phi_L \) gradients are larger in regions with high positive curvatures (cliff) compared with that in regions with negative curvatures (pool). However, the difference is reduced in cases B and C. It is also noted that the HRR increases with increasing \( \phi_L \) gradient and the HRR is stronger in positive curvature regions. However, both the peak heat release rate and largest \( \phi_L \) gradient do not happen in regions with peak curvatures.
In the 2D slices of Fig. 6.5, the contour of H mass fraction agrees well with the contour of HRR. In experimental studies, the heat release regions cannot be measured directly, whereas usually be marked by chemical radicals. Therefore, it is necessary to examine if H could be used as a reliable HRR marker in high-pressure lean flames with varying equivalence ratios. Figure 6.14 shows the distribution of H mass fraction against HRR, which is coloured by local temperatures.

For the ultra-lean flame, H mass fraction shows a good spatial correlation with HRR. When the equivalence ratio increases, the correlation between $Y_H$ and HRR is becoming poor with a broader distribution area. In flame $\phi = 0.6$, it even shows two scatter branches which is separated by $T \approx 1550K$. Therefore, these results show that radical H could be directly used as an HRR marker under ultra-lean conditions for turbulent flames at high pressures. Under slight-lean conditions, the marker performs very bad.

It is interesting to note that the temperature increases in the clockwise direction for all three flames in Fig. 6.14, which indicates that the HRR is determined by both H mass fraction and local temperature. Therefore, Eq. 5.3 is introduced to predict the HRR using both H mass fraction and temperature for flames with varying equivalence ratios.
Fig. 6.15 Scatter plots of scaled H mass fraction and heat release. The scatters are coloured by local temperatures.

When $n = 3.6$, the HRR distribution against $Y_H^*$ is shown in Fig. 6.15. A striking observation is that the scaled H mass fraction shows a linear spatial correlation with HRR in all three high-pressure flames with different equivalence ratios. For the case $\phi = 0.6$, the degree of fitting is very high even though the distribution is slightly broader than that of the other two cases. Therefore, H could be used as a perfect HRR marker in high-pressure flames with varying equivalence ratios when it is scaled by temperature.

6.4 Response of elementary reactions to equivalence ratio variation

As a multi-step chemical mechanism is adopted in this study, the heat release we mentioned above is the overall presentation of all elementary reactions. When the equivalence ratio varies, the radical distribution and elementary reaction must be affected. Studying the heat release from main elementary reactions will extend our understanding of premixed flames. There are 21 reversible reactions in the used mechanism and the five elementary reactions listed in Table 4.3 contribute to ~90% of the total heat releases.
Firstly, Fig. 6.16 presents the heat release contributions of main reactions to the integrated heat release for the high-pressure turbulent flames with equivalence ratios 0.4, 0.5 and 0.6, respectively.

It is observed that the chemical pathways are globally modified. With increasing equivalence ratio, there are significant changes for R8 and R13. The heat release contribution of R8 increases by 7.5%, while the contribution of R13 decreases by 7.8% when the equivalence ratio increases from 0.4 to 0.6. For R9 and R11, the change of contribution is around 3.5%; the contribution of R3 increases slightly.

Secondly, Fig. 6.17 presents the average heat release rate of the selected 5 elementary reactions versus temperature in the reaction zones of turbulent flames at different equivalence ratios. As expected, the elementary reactions show high HRR above the adiabatic temperature in case \( \phi = 0.4 \) and 0.5, which will contribute to the hot spots in the reaction zones. Before reaching the adiabatic temperature, every reaction exhibits an HRR peak. It is noticed that the HRR peak arises from 1200 K for all three
cases, while the slope is decreasing with increasing equivalence ratio. Moreover, the heat release under moderate temperatures (600-1200 K) is significantly enhanced when the equivalence ratio is increased from 0.4 to 0.6, especially for the dominating reactions R9 and R13. In the high-temperature regions ($T > 1200$ K), the heat release contributions from R3 and R8 are more prominent with increasing equivalence ratios. R3 overtakes R13 as the second most dominating reaction in case $\phi = 0.6$.

As shown in Eqs. 3.23 and 3.31, two key factors, rate constant $K$ and species concentration $X_k$, directly determine the reaction rate. Thus, the species concentrations and rate constants as a function of temperature are presented in Figs. 6.18 and 6.19 to assist the discussion of heat release in reaction zones. Overall, it is seen in Fig. 6.18 that the fractions of all species are obviously increased with increasing equivalence ratio, which can explain the enhanced heat release rate under high $\phi$. When the temperature is higher than adiabatic temperature, it is noted that there exist considerable radicals for cases $\phi = 0.4$ and 0.5, which will lead to the formation of hot spots in the reaction zone. This observation is consistent with the $\phi_L$ plateaus in Fig. 6.8. Although differential diffusion is expected to be less important under high Ka [36], the effects are still considerable when the equivalence ratio is increased in this study. From Fig. 6.18, it is
6.4 Response of elementary reactions to equivalence ratio variation

It is evident that the diffusion of H and OH is enhanced under high $\phi$, leading to a broader and uniform distribution.

Fig. 6.18 Scatter plots of main species mole fractions in the reaction zone. The solid red lines denote the mean values.
Effects of Equivalence Ratios in High-Ka Lean Premixed H2/air Flames at High Pressures

Fig. 6.19 Forward and reverse reaction rate constants versus temperature for the main heat release reactions. The solid lines denote the forward reaction rate and the dashed lines denote the reverse reaction rate.

In Fig. 6.19, it is seen that the rate constants of reverse reactions are negligible compared with that of forward reactions, despite the relatively high rate constant of reverse reaction R3 under high temperature. Therefore, in the following discussion of elementary reactions, we only consider the positive reactions.

For R9 and R13, the positive reaction rates show exponential decay with increasing temperature. Moreover, the mole fractions of H and OH are increased in moderate-temperature regions for case \( \phi = 0.6 \). As a result, the heat release is enhanced in moderate temperatures for case \( \phi = 0.6 \) in Fig. 6.17. For reaction 3, the positive reaction rate and OH mole fraction all experience exponential growth with increasing temperature, which is more significant under higher equivalence ratio. These dual effects promote the reaction of R3 under high \( \phi \), which explains why R3 overtakes R13 as the second most dominating reaction for case \( \phi = 0.6 \). It is also noted in Fig. 6.17
that R8 is enhanced in high-temperature regions under high $\phi$. This could be attributed to the high contribution of H$_2$O to third-body [M] in Eq. 3.31. When the equivalence ratio is increased, the H$_2$O concentration under high temperatures is also increased, which will lead to a high [M] value. For R11, the positive rate constant levels off after 1000 K. Thus, although the HRR of R11 is increased, the difference between moderate-temperature and high-temperature regions is not as significant as other reactions.

6.5 Summary

The characteristics of heat release under various equivalence ratios ($\phi = 0.4, 0.5, 0.6$) are investigated by three-dimensional direct numerical simulations of lean H$_2$/air flames at same Ka under 20 atm. With increasing equivalence ratio, the heat release rate is significantly enhanced and the reaction zone is broadened. However, the flame fronts show similar topological structures in the flames with different equivalence ratios. Under ultra-lean condition, the local equivalence ratio ($\phi_L$) is found to be significantly higher than the mixture equivalence ratio, and the gradient of $\phi_L$ is obviously increased. As a result, hot spots with temperatures higher than the adiabatic temperature are observed in the reaction zone. It indicates that the benefits of lean combustion in term of lowering peak flame temperature may be slightly cut down under ultra-lean conditions. Moreover, local chemical pathways are modified in different temperature windows when the mixture equivalence ratio varies.
Chapter 7

Effects of Integral Length Scales in Lean Premixed H₂/air flames

One important question relevant to DNS studies is the selection of the ratio of integral length scale to laminar flame thickness ($l_t/\delta_L$). It can be seen from Fig. 2.4, in most cases, the $l_t/\delta_L$ ratio is set close to unity. The main reason is that computational cost increases exponentially as the $l_t/\delta_L$ ratio increases. However, in experimental and industrial combustion devices, the integral length scale is over ten times the laminar flame thickness [21, 183]. In turbulent flames, the integral length scale could affect local flame stretch, and then further alter local temperature and species distribution [4]. Thus, it is necessary to examine whether the flame structures and chemical processes are fundamentally modified by the integral length scale. In other words, are DNS results with $l_t/\delta_L \approx 1$ representative? Moreover, when we investigate the effects of pressure on turbulent flames, the $l_t/\delta_L$ ratio and Karlovitz number are usually kept constant to isolate pressure effects from the other parameters [108, 184]. It is also important to know whether this approach is reasonable for flames with varied Karlovitz numbers. This chapter aims to investigate the effects of integral length scales on turbulent flame
structures and chemical reactions under different pressures. The two problems above are emphasized in the discussion of turbulent lean premixed H₂/air flames.

7.1 Simulation details

7.1.1 Simulation parameters

Table 7.1 Simulation parameters of turbulent flames with different integral length scales.

<table>
<thead>
<tr>
<th>Case</th>
<th>5PLhalf</th>
<th>5PL1</th>
<th>5PL2</th>
<th>1PLhalf</th>
<th>1PL1</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P$ (atm)</td>
<td>5</td>
<td>5</td>
<td>5</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$S_L$ (cm/s)</td>
<td>6.53</td>
<td>6.53</td>
<td>6.53</td>
<td>23</td>
<td>23</td>
</tr>
<tr>
<td>$\delta_L$ (cm)</td>
<td>0.0315</td>
<td>0.0315</td>
<td>0.0315</td>
<td>0.06</td>
<td>0.06</td>
</tr>
<tr>
<td>$u'$ (cm/s)</td>
<td>119</td>
<td>150</td>
<td>189</td>
<td>150</td>
<td>478</td>
</tr>
<tr>
<td>$l_t$ (cm)</td>
<td>0.01575</td>
<td>0.0315</td>
<td>0.063</td>
<td>0.0315</td>
<td>0.06</td>
</tr>
<tr>
<td>$v$ (cm²/s)</td>
<td>0.0362</td>
<td>0.0362</td>
<td>0.0362</td>
<td>0.0181</td>
<td>0.0181</td>
</tr>
<tr>
<td>$l_t/\delta_L$</td>
<td>0.5</td>
<td>1.0</td>
<td>2.0</td>
<td>0.525</td>
<td>1.0</td>
</tr>
<tr>
<td>$u'/S_L$</td>
<td>51.2</td>
<td>23.0</td>
<td>28.9</td>
<td>6.52</td>
<td>20.78</td>
</tr>
<tr>
<td>$\delta_L/\Delta x$</td>
<td>51</td>
<td>38</td>
<td>19</td>
<td>37</td>
<td>26</td>
</tr>
<tr>
<td>$\Delta x/\eta$</td>
<td>0.75</td>
<td>1.0</td>
<td>2.0</td>
<td>0.6</td>
<td>1.74</td>
</tr>
<tr>
<td>Re</td>
<td>52</td>
<td>131</td>
<td>329</td>
<td>26</td>
<td>158</td>
</tr>
<tr>
<td>Ka</td>
<td>262</td>
<td>262</td>
<td>262</td>
<td>63</td>
<td>262</td>
</tr>
</tbody>
</table>

There are five DNS cases in this chapter and the parameters of these cases are listed in Table 7.1. In all cases, the temperature of the fresh mixture is 298 K and the equivalence ratio is $\varphi = 0.4$. Cases 5PLhalf, 5PL1 and 5PL2 are characterised by the same pressure (5 atm) but different ratios of integral length scale to laminar flame thickness. Case 1PLhalf is initialised with an identical turbulence field as case 5PL1 but pressure is kept at 1 atm. To isolate the effects of pressure from other parameters,
case 1PL1 is performed at the atmospheric pressure while the Karlovitz number and the ratio of integral length scale to laminar flame thickness are identical to that of case 5PL1. For the three cases at 5 atm, the properties of laminar flames are the same, whereas the ratio of integral length scale to laminar flame thickness increases from 0.5 to 2.0. When pressure is decreased from 5 to 1 atm, the laminar flame speed increases from 6.53 cm/s to 23 cm/s, and the flame thickness increases from 0.0315 to 0.06 cm. Therefore, the ratio of integral length scale to laminar flame thickness in case 1PLhalf decreases to 0.525. As the kinematic viscosity is dramatically reduced under the elevated pressure, the RMS velocity in case 1PL1 is increased to 478 cm/s to ensure the same Ka as that of case 5PL1.

Recalling the relations derived by Peters [48], case 1PLhalf is located in the thin reaction zone, and the other four cases are situated at the boundary of the thin reaction zone and the broken reaction zone, which is demonstrated in Fig. 7.1.

![Fig. 7.1 Turbulent combustion regime diagram with locations of current simulation cases.](image)

**7.1.2 Grid-independence test**

In all cases, there are 20 integral length scales across the domain length. For cases 5PLhalf and 1PL1, the grid resolution is 512×256×256; for cases 5PL1 and 5PL2, the
grid resolution is $768 \times 384 \times 384$; for case 1PLhalf, the grid resolution is $384 \times 192 \times 192$. It can be seen from Table 1, there are 19-51 grids across the laminar flame thickness and the ratio of grid size to Kolmogorov length scale is kept under 2.0, which is sufficient to resolve flame and turbulence structures in DNS with detailed chemistry [35]. As in previous chapters, two-dimensional (2D) DNS cases were also performed to optimise the selection of simulation parameters. Figure 7.2 shows the temporal evolution of integrated heat release rate (HRR) and Fig. 7.3 shows the PDF of normalised HRR.

Fig. 7.2 Temporal evolution of integrated heat release rate for cases 5PLhalf and 5PL2 and the corresponding grid-independence test cases.

Fig. 7.3 Probability density function of normalised heat release rate for cases 5PLhalf and 5PL2 and the corresponding grid-independence test cases at $t / \tau_l = 4$. 
It demonstrates that there is a good agreement in the temporal evolution of integrated heat release between the baseline case and the refined case. The small fluctuations for the PDF distributions are negligible. Therefore, current turbulent flames are well-resolved, and the simulations are grid independent.

7.2 Effects of integral length scales on flame structures under 5 atm

Figure 7.4 shows the 2D snapshots of heat release rate for the three cases under 5 atm with different $l/\delta_L$ ratios (0.5, 1.0 and 2.0) at the same large eddy turn-over time. The dashed lines bound the regions with HRR higher than the peak HRR of the corresponding laminar flame (HRR$_{1D,peak}$). To illustrate the reaction zone structures, the three plots are presented with the real domain size instead of scaled size. For the flame with $l/\delta_L = 0.5$, the reaction zone is continuous and there are tiny regions with HRR higher than HRR$_{1D,peak}$. When the $l/\delta_L$ ratio is increased to 1.0 and 2.0, the flame front is seriously wrinkled and there exists long narrow regions with HRR higher than HRR$_{1D,peak}$. Comparing the three flames, the HRR is dramatically enhanced when the $l/\delta_L$ ratio is increased from 0.5 to 1.0, whereas the enhancement is not apparent when the ratio is further increased to 2.0.

The stretched flame cells indicate that the scale of flame structures is comparable with the corresponding integral length scale. Flame 5PLhalf shows laminar flame characteristics even though it is located at the boundary of the thin reaction zone and the broken reaction zone. It was discussed by Meneveau and Poinsot [86, 87] that the lifetime of small vortices is very short and they are unable to stretch the flame front when the $l/\delta_L$ ratio is lower than unity. The conversion of vortex strain to flame stretch
is dramatically reduced when the $l_0/\delta_L$ ratio decreases. In flames 5PL1 and 5PL2, the convex region toward the unburnt gas is larger than the concave region protruding to the burnt gas. The heat release rate in the convex regions is higher than that in the concaves regions, which is mainly induced by the concentration of reactant and defocusing of heat in convex regions [185].

![Fig. 7.4 2D snapshots of heat release rate for cases 5PLhalf (a), 5PL1 (b) and 5PL2(c). The dashed lines denote the peak heat release rate of the corresponding laminar flame at 5 atm.](image)

The reaction zone is bounded by $c = 0.01$ and 0.99. Figure 7.5 demonstrates the instantaneous streamlines coloured by temperature contours near the reaction zone, which is also presented with the real domain size as that in Fig. 7.4. The three cases are characterised by $l_0 \approx \eta < \delta_L$, so that small vortices could be found in the reaction zone. The size of large vortices is comparable with the corresponding integral length scale. Nevertheless, no Kolmogorov-scale vortices are observed as they are quickly dissipated. It can be seen from Fig. 7.5 (a), there is no apparent change in reaction zone thickness across the domain height in flame 5PLhalf. However, the reaction zone is significantly wrinkled, and the local thickness of reaction zone is dramatically increased or reduced.
when the $l/\delta_L$ ratio is increased to 1.0. This phenomenon becomes more evident in flame 5PL2. Obvious compression in convex regions and expansion in concave regions are observed. For example, the reaction zone thickness in the concave region around $x = 0.6$ cm and $y = 0.4$ cm almost doubles, while the thickness around $x = 0.6$ and $y = 0.55$ is less than half of that in flame 5PLhalf. The magnitude of progress variable gradient is usually denoted as the surface density function (SDF) [185]. The observation from Fig. 7.5 indicates that SDF is locally changed when $l/\delta_L \geq 1$.

![Instantaneous streamlines coloured by temperature contours near the reaction zones of flames 5Plhalf (a), 5PL1 (b) and 5PL2(c).](image)

Furthermore, the flame thickness across the preheat layer, inner layer and oxidation layer are examined to investigate the effects of $l/\delta_L$ ratio on turbulent flame structures. The average progress thickness is quantified by:

$$\Delta_T = \frac{\int_{T_{\text{g}}-50<T<T_{\text{b}}+50} V_s \, \mathrm{d}S}{S}$$

(7.1)
where \( V \) is the iso-volume with temperature in the range \( T_0 - 50 < T < T_0 + 50 \) and \( S \) is the cross-sectional area of the computational domain. The average progress thickness is scaled by the corresponding progress thickness of the laminar flame \( \Delta_L \) at 5 atm.

Fig. 7.6 shows the scaled progress thickness and iso-temperature surface area for the three turbulent flames at 5 atm. It is noted that the flame with \( l/\delta_L = 0.5 \) is only slightly thickened in the low-temperature and high-temperature regions. When the \( l/\delta_L \) ratio is increased, the flame is significantly thickened in the reaction zone, and the extent is positively correlated with the \( l/\delta_L \) ratio, even though there is an obvious drop at 1350 K in case 5PL2. Figure 7.6 (b) presents the iso-temperature areas across the flame. In flame 5Plhalf, the scaled surface area is only slightly enlarged in low-temperature regions and then drops close to unity as the temperature increases. The flame thickness and flame front area are quantitatively close to those of laminar flames. It is evident that the area is less sensitive to the temperature and it is proportional to the \( l/\delta_L \) ratio. These findings imply that when the integral length scale is smaller than the laminar flame thickness, turbulence lacks the capability to interrupt the reaction zone and affect the scalar mixing even though the Karlovitz number is high.
To statistically examine the effects of integral length scales on turbulent flames structures, Fig. 7.7 presents the PDFs of mean curvature and tangential strain rate on the flame fronts. The mean curvature is given by Eq. 4.10. The tangential strain rate is given by:

\[
a_t = (\delta_{ij} - n_in_j)\tilde{S}_{ij} = (\delta_{ij} - n_in_j)\frac{\partial U_i}{\partial x_j}
\]

where \(\delta_{ij}\) is the Kronecker delta, \(n_i\) and \(U_i\) are respectively the components of normal unit vector and velocity in \(i\) direction.

The results shown in Fig. 7.7 are calculated over the iso-surfaces \(c = 0.7, 0.8\) and \(0.9\). It is surprising to note that flame 5PLhalf exhibits comparable distributions of curvature and strain rate compared with the other two cases despite the flame presents laminar characteristics in Figs. 7.4-7.6. The curvature distribution is symmetric and the average value is close to zero in flame 5PLhalf. When the \(l/\delta_L\) ratio is increased to 1.0, the PDF distribution is slightly skewed toward the positive values. In flame 5PL2, the probability of finding negative curvatures is obviously decreased, and the mean curvature value is positive. It is known that the high local curvature is mainly induced
by vortices smaller than the flame thickness [87]. As the three flames are characterised by the same Kolmogorov scale, they show similar probability of high curvatures. However, the probability of low curvatures in flame 5PL2 is significantly higher than that in the other two cases.

In Fig. 7.7 (b), the mean strain rate is positive, which agrees with the finding in Refs. [87, 185]. When the integral length scale is smaller than the laminar flame thickness, the mean strain rate is lower, and the vortices are unable to enlarge the flame surface even though the positive strain rate is higher. It is also noted that the probability of finding positive strain rate increases with increasing $l/\delta_L$ ratio even though the effect is not obvious when the integral length scale is larger than the flame thickness. When the $l/\delta_L$ ratio increases from 0.5 to 1.0, the probability of negative strain rate and small positive strain rate decreases and the probability of high positive strain rate increases. In flame 5PL2, the probability of low and moderate positive strain rate is significantly higher than the other cases.

### 7.3 Effects of integral length scales on flame structures under different pressures

In turbulent flame simulations under different pressures, the $l/\delta_L$ ratio is usually kept constant to isolate the effects of pressure from the other dominating parameters [108]. As a result, the effects of integral length scales are not yet clarified when pressure changes. Based on the above discussion, this section is trying to identify the effects of integral length scales and pressure on turbulent flame structures. Figure 7.8 shows the instantaneous streamlines coloured by temperature contours near the reaction zone. Cases 1PLhalf and 5PL1 are presented with the absolute domain size, while case 1PL1
7.3 Effects of integral length scales on flame structures under different pressures

is demonstrated with the size scaled by the corresponding domain length. The three flames show comparable vortex sizes in the reaction zone. As expected, the absolute flame thickness at 1 atm is larger than that at 5 atm. In flames 1PLhalf and 5PL1, the turbulent eddies are dramatically dissipated by viscosity in the high-temperature regions and there exhibit laminar characteristics after the flame front. Nevertheless, turbulent structures are observed after the reaction zone in flame 1PL1. It indicates that the flow structures after the reaction zone are more relevant to the turbulence intensity $u'$ instead of $K_a$.

![Fig. 7.8 Instantaneous streamlines coloured by temperature contours near the reaction zones of cases 1Plhalf, 1PL1 and 5PL1](image)

For flame 1PLhalf, the integral length scale is smaller than the flame thickness, thus the vortices are unable to effectively stretch the flame front even though there are many turbulent eddies in the reaction zone. While for flame 1PL1 and 5PL1, the reaction zones are obviously wrinkled and the scaled thickness is almost identical. The reaction zones show similar SDF values. To further examine the reaction zone structure, Fig. 7.9 demonstrates the average progress thickness and iso-temperature surface area for the flames at different pressures. It is noted that flame 1PLhalf is only slightly thickened in the reaction zone and the scaled surface area is close to unity. This finding
agrees with that of flame 5PLhalf, and it confirms that the vortices are unable to modify flame structures when the integral length scale is smaller than the flame thickness. For the two flames with the same Ka but under different pressures, the flame thickness and flame front area are significantly increased by turbulence, whereas different changing trends are clearly observed across the reaction zone.

![Fig. 7.9 Scaled flame thickness (a) and flame surface area (b) versus temperature for cases 1PLhalf, 1PL1 and 5PL1, respectively.](image)

In flame 1PL1, the relative thickness increases slowly from 1.3 to 1.5 as the temperature increases from 400 to 1000 K, after that the relative thickness starts to decrease until it reaches unity at 1350 K. For flame 5PL1, the relative thickness is stable below 800 K and then increases sharply to 2.2 at 1350 K. Correspondingly, the relative area decreases with increasing temperature in flame 1PL1 while it increases as the temperature increases. It can be explained by the effects of pressure on the thickness of reaction zones. Peters [48] argued that the ratio of inner layer thickness to flame thermal thickness decreases as pressure is elevated. In the studied cases, the Kolmogorov scale is larger than the inner layer thickness, and the small eddies cannot penetrate into and disturb the inner layer. However, vortices can change the flame structures before the inner layer. At 1 atm, the temperature corresponding to the peak HRR in laminar flame
is around 1100 K, while the value is about 1300 K at 5 atm. Therefore, the relative thickness is obviously decreased when the temperature is higher than 1100 K in flame 1PL1. Nevertheless, no decrease is observed in flame 5PL1 after 1300 K. It may be attributed to the shrunken reaction layer toward high temperatures, and the selected temperature range (1250 K < T < 1350 K) is too large to capture the inner layer thickness under 5 atm.

Figure 7.10 presents the PDFs of mean curvature and tangential strain rate for the three flames. It is observed that the distributions for flame 1PL1 overlap with those of 5PL1, which indicates that keeping constant \( \frac{l}{\delta_L} \) ratio and \( K_a \) to isolate pressure effects in turbulent flames is reliable. For all three flames, the curvature distribution is symmetric. In flame 1PL1 and 5PL1, the Kolmogorov scale is smaller than that in flame 1PLhalf, so that there is a higher probability of finding high curvatures. In our previous study [184] of turbulent flames with unity \( \frac{l}{\delta_L} \) ratio, it was demonstrated that the positive curvature increases as pressure is elevated. However, it is not evident in the present cases. In another study [133] of turbulent flames initialised with an identical turbulent box, the \( \frac{l}{\delta_L} \) ratio is higher, and the probability of finding positive curvatures...
is increasing when pressure is elevated. Combined with Fig. 7.7, it indicates that the probability of finding positive curvatures is co-determined by pressure and $l/\delta_L$ ratio. The effects of $l/\delta_L$ ratio are more significant in the studied cases.

In Fig. 7.10 (b), the mean strain rate is positive and increases as the Ka increases, which is consistent with previous findings from DNS studies [96]. The high strain rate in high Ka cases is associated with the small Kolmogorov scales [86]. Combined with the results in Fig. 7.12, it is noted that the positive strain rate in convex regions is higher in flame 5PL1 than that in flame 1PL1, which indicates that the convex regions could be highly stretched under elevated pressures. This finding directly explains why finger structures toward the unburnt gas were formed in the lean premixed turbulent flame under elevated pressures in our previous DNS results [133].

7.4 Identification of heat release rate and chemical pathways

To save computational resources, most of 3D DNS studies of turbulent flames are performed with small integral length scales [36, 39, 40]. However, the $l/\delta_L$ ratio is relatively high in experimental studies [21, 183]. The altered flame stretch by $l/\delta_L$ ratio could modify local temperature and radical distribution [4]. It is necessary to examine whether heat release rate and chemical pathways are fundamentally modified by the integral length scale. In Fig. 7.4, it demonstrates that the peak HRR in turbulent flames does not change significantly when the $l/\delta_L$ ratio is higher than unity. To further examine the performance of heat release, Fig. 7.11 shows the joint PDF of scaled heat release rate against the progress variable across the whole flame.

At 5 atm, the HRR in the flame with $l/\delta_L = 1$ is enhanced to the same extent as that in the flame with $l/\delta_L = 2$, even though the distribution is broader under the higher ratio.
As expected, flame 5PLhalf shows an obviously different distribution. The peak HRR is less than half of that in flames 5PL1 and 5PL2, and the heat release in the region \( c < 0.7 \) is marginal. The results indicate that when the \( l_t/\delta_L \) ratio is higher than unity, the turbulent flame could capture the main features of heat release as that in flames with higher \( l_t/\delta_L \) ratios.

![Fig. 7.11 Scatter plots of scaled heat release rate versus progress variable in turbulent flames with different \( l_t/\delta_L \) ratios and pressures.](image)

Under the atmospheric pressure, the scatter distribution is extended to regions with low values of progress variable, which is more evident in the high Ka flame. In flame 1PL1, the progress variable value corresponding to the peak heat release rate is around 0.6, whereas the value is 0.85 in flame 5PL1. This is directly induced by the shrunk inner layer under elevated pressures. Even though the two flames 1PL1 and 5PL1 are characterised by the same Ka, the enhancement of HRR is significantly higher at elevated pressures. It was indicated by Poinsot et al. [87] that HRR is positively
correlated with flame stretch. The flame stretch is expressed as \( K = a_f + S_d \nabla \cdot \vec{n} \), where \( S_d \) is the displacement speed and it is proportional to \( A_r / A_r \) [97]. As demonstrated in Figs. 7.9 and 7.10, the tangential strain rate and curvature are identical in the two flames, while the scaled surface area in flame SPL1 is larger in the reaction zone. Therefore, the heat release is enhanced in regions with high values of progress variable for flame SPL1.

Fig. 7.12 Scatter plots of scaled heat release rate and mean curvature on the flame fronts coloured by tangential strain rate.

Figure 7.12 gives the scatter plots of heat release rate and mean curvatures on the flame fronts. The scatter is coloured by the corresponding tangential strain rate. For all five flames, it is noted that HRR in regions with positive curvatures is higher than that in regions with negative curvatures, and it becomes more evident when the \( l_0/\delta_L \) ratio is larger than unity, which is consistent with the HRR contours in Fig. 7.4. In the high Ka flames at 5 atm, the local regions with high tangential strain rate are also characterised by high HRR, whereas it is not evident for the high Ka flame at atmospheric pressure.
Moreover, the tangential strain rate reaches peak values near zero curvature regions and it decreases with increasing $\left|\kappa\alpha\right|$, which was also reported by Chakraborty et al. [185] for turbulent H$_2$/air flames. In positive-curvature regions, the probability of finding high negative tangential strain rate is higher than that in negative-curvature regions. In summary, the main characteristics of curvature and tangential strain rate on flame front could be observed when $l_t/\delta_L \geq 1$ and the effects of turbulence on flame structures are modified by elevated pressures.

Fig. 7.13 Heat release contributions of elementary reactions against $l_t/\delta_L$ ratios for turbulent flames with the same pressure but different $l_t/\delta_L$ ratios (a) and turbulent flames under different pressures (b). The dashed lines in (b) correspond to the heat release contributions in laminar flames. The mark $\times$ denotes the heat release contributions in flame 1PLhalf.

To examine the effects of integral length scales on chemical pathways, Fig. 7.13 presents the heat release contribution of seven main reactions from the chemical mechanism. The data of laminar flames is also given as a baseline. At 5atm, the elementary reactions are slightly promoted or suppressed when the $l_t/\delta_L$ ratio increases from 0.5 to 1. When the ratio is further increased to 2, the change is marginal, which indicates that the effects of integral length scale on the modification of global chemical pathways are negligible, especially when $l_t/\delta_L \geq 1$. This finding is also observed in the
turbulent flames at the atmospheric pressure in Fig. 7.13 (b). Comparing flames 1PL1 and 5PL1, it indicates that the effects of pressure on the modification of global chemical pathways are more significant than that of the integral length scale. The maximum change happens for reaction $\text{HO}_2 + \text{OH} = \text{H}_2\text{O} + \text{O}_2$ in the laminar flames, with a 14% enhancement when pressure is increased from 1 atm to 5 atm. However, the promotion is reduced by 6% in the turbulent flames.

### 7.5 Summary

Five DNS cases with different ratios of integral length scale to laminar flame thickness ($l_l/\delta_L$) are performed under 1 atm and 5 atm to investigate the effects of the integral length scale and pressure on turbulent flame structures and chemical reactions. When the integral length scale is smaller than the thickness of the corresponding laminar flame, the reaction zone shows laminar flame characteristics. Turbulence is unable to stretch and interrupt the reaction zone. However, the distributions of curvature and tangential strain rate are comparable with those in flames with $l_l/\delta_L \geq 1.0$ when the Karlovitz number is fixed at a high value. It is found that keeping constant $l_l/\delta_L$ ratio and $K_a$ could isolate the effects of pressure on flame front structures. Moreover, global chemical pathways are insensitive to the $l_l/\delta_L$ ratio but sensitive to pressure. The turbulent flame with unity $l_l/\delta_L$ ratio could capture the main features of heat release as those in flames with higher $l_l/\delta_L$ ratios.
Chapter 8

Conclusions and Future Work

In this thesis, a fundamental study has been conducted of turbulent premixed flames under conditions relevant to gas turbines. A series of three-dimensional direct numerical simulations (DNS) have been performed for lean premixed H$_2$/air flames at high Karlovitz numbers under elevated pressures. A 9-species 21-reaction chemical mechanism is adopted in the simulations. The effects of Karlovitz number, pressure, equivalence ratio and integral length scale are investigated by analysing flame structures and chemical pathways.

8.1 Conclusions

8.1.1 Effects of Karlovitz number

The effects of the Karlovitz number on flame structures and chemical pathways have been analysed by direct numerical simulation of lean premixed turbulent H$_2$/air flames at 5 atm in the thin reaction zone and the distributed reaction zone. The domain size and the integral length scale are kept constant to isolate the effects of Ka from the other controlling parameters. Statistical analysis indicates that the effects of Ka on turbulence-flame interactions and the key phenomena of turbulent flames could be clearly observed at two large eddy turnover times.
Due to the rapid dissipation of small-scale vortices, the flame with a low Ka (P5_L) shows analogous structures with the flame of a moderate Ka (P5_M). The flame fronts of the two cases are highly stretched by large vortices with a lack of sharp edges. For the flame at the high Ka (P5_H), the sizes of small eddies are dramatically reduced, and a significant number of small cusps and pockets are formed on the flame front. Although some small eddies may be dissipated by viscosity, the existing small vortices are sufficiently energetic to disrupt the reaction zone and even the inner layer at a high Ka.

It is observed that the flow shows laminar characteristics of regular streamlines after the flame front in cases P5_L and P5_M, whereas both small and large vortices could survive after the flame front in flame P5_H. Moreover, the distribution of radical H matches well with the distribution of heat release rate, which indicates that the atomic hydrogen could be used as a reliable HRR marker for flames with different turbulence intensities at elevated pressures.

When the Karlovitz number increases, even though hydrogen diffusion effects are reduced, the turbulence-flame interaction is still significantly affected by differential diffusion. Statistical analysis of the flame fronts identifies that the probability of high curvatures (both positive and negative) increases with increasing Ka, while the asymmetry of the positive and negative curvature PDFs is insensitive to turbulence intensity. The scatter of heat release rate becomes widely distributed at high Ka, which provides a sign of transition to distributed burning. Regions with high positive curvatures are characterised by relatively low heat release rates compared with regions with high negative curvatures under the same H2 consumption rate. Moreover, the flame has different effects on turbulence with low or high intensities. In flame P5_L, the iso-
surface area is becoming larger with increasing progress variable, while the opposite
trend is observed in flame P5H.

Although the effects of the Karlovitz number on global fractional contribution of
elementary reactions are limited, chemical pathways can be modified locally. Overall,
the heat release of an individual reaction is promoted in regions with low values of
progress variable and suppressed in regions with high values of progress variable.
Moreover, the local heat release contributions are varied in different reactions when Ka
increases. Compared with flame P5L, the local contributions from R3 and R9 in flame
P5H increase obviously in the region $0.5 < c < 0.9$, whereas the local contributions
from R9 and R13 decrease slightly. The changed distributions of heat release result
from the altered species distributions by turbulence. The enhanced scalar mixing
increases the radical concentrations ($Y_H$, $Y_O$, $Y_{OH}$ and $Y_{HO2}$) in regions with low values
of progress variable. As a result, the elementary reactions are promoted, and the reaction
zone is extended to lower temperature regions.

8.1.2 Effects of pressure

The effects of pressure on flame structures and chemical pathways are investigated
by DNS of lean premixed turbulent H2/air flames at 1, 5 and 20 atm. To isolate the
effects of pressure, two sets of cases are performed under different pressures. The first
set is characterised by a fixed high Ka and the second set is initialised with an identical
turbulent field.

In laminar flames, the thickness of the heat release zone is decreasing with
increasing pressure, even though the peak heat release rate is dramatically high at high
pressures. In turbulent flames, the reaction region is becoming discontinuous and
getting narrower when pressure is elevated. At a high Ka, all the flame fronts are severely wrinkled. Under 1 and 5 atm, the sizes of small flame cells are dramatically reduced, and small eddies are observed after the high-temperature reaction zone. However, there are no small vortices after the flame front for the same Ka flame at 20 atm, even though the flame edges are quite sharp. Comparing the cases initialised with an identical turbulent field, it is found that eddies are quickly dissipated at low pressures. Case S1 even shows laminar characteristics of regular streamlines after the flame front.

It is noted that flame structures for cases with the same Ka but different pressures are statistically different. The probability of high curvatures is increasing with decreasing pressures. The approximation of unity Schmidt number (Sc = 1) in theoretical analysis introduces errors in the estimation of the smallest flow thickness and the Karlovitz number. These findings suggest the necessity for modification of Peters’s regime diagram at elevated pressures, which is an interesting area for future investigation. Moreover, it is found that the relative probability of positive curvature to negative curvature is sensitive to pressure, but insensitive to turbulent intensities. The decorrelation degree of heat release and H₂ consumption is directly related to the turbulence intensity \( u' \), and the decorrelation reduces with increasing pressure. It is interesting to find that under the same H₂ consumption rate, heat release rates in regions with high-positive curvatures are relatively low compared with areas with high-negative curvatures. However, this phenomenon disappears in the flame at 20 atm.

Pressure could globally modify chemical pathways in both laminar and turbulent flames, and the effects are more significant than that of turbulence intensity and the Karlovitz number. At 1 atm, the differences in fractional heat release between laminar flames and turbulent flames are small. However, it becomes significant at 20 atm. It is
also found that the effects of pressure on modification of local chemical pathways are significant. When pressure is elevated, the heat release region is shrinking with a narrow progress variable range, and the heat release intensity is increasing. Moreover, the fractional consumption/production of radicals in elementary reaction is dramatically modified by pressure. At elevated pressures, chemical pathways are susceptible to turbulence in high Ka flames.

8.1.3 Effects of equivalence ratio

Three DNS cases with equivalence ratios of 0.4, 0.5 and 0.6 are performed under conditions relevant to gas turbines. The Karlovitz number, the ratio of domain length to integral length scale and the ratio of integral length scale to laminar flame thickness are kept constant to isolate the effects of equivalence ratio from other controlling parameters. Reaction zone structures, local equivalence ratios, flame front curvatures and elementary reactions are examined statistically with the objective of extending our understanding of lean combustion under high-pressure and high-Ka conditions.

Under different lean conditions, the turbulent flame fronts show similar topological structures for the three cases under the same Karlovitz number, even though the higher $\phi$ case is characterised by stronger heat release. It is also noted that the relative thickness of reaction zone ($\delta_r/\delta_L$) increases for both laminar and turbulent flames with increasing equivalence ratio. However, the extent of increase in heat release and reaction layer thickness is reduced under high $\phi$, which indicates that turbulent transport is overtaking differential diffusion as the main effect on the high Ka flames.

Turbulent flames behave differently in positive and negative curvature regions. It is observed that the trenches of local equivalence ratio $\phi_L$ are located at concave
structures outside the reaction zone, while $\phi_L$ plateaus are situated at convex structures inside the reaction zone. The $\phi_L$ gradients are larger in regions with high positive curvatures compared with that in regions with negative curvatures. With increasing equivalence ratio, the difference of gradients in convex and concave regions reduces, which is primarily due to the decreased Lewis number. As a result, the trench ‘depth’ and plateau ‘height’ are reduced. In addition, $\phi_L$ is found to be significantly higher than mixture $\phi$ under ultra-lean conditions, resulting in hot spots in the reaction zone. This finding suggests that, for pollutant control, it is not “the leaner the better”, and this is instructive for industrial applications. Furthermore, it is observed that the radical H could be directly used as an HRR marker under ultra-lean conditions, while it performs poorly under slight-lean conditions. Nevertheless, the H mass fraction shows a linear spatial correlation with HRR in all three flames with different equivalence ratios when it is scaled by temperature.

The heat release behaviour in the reaction zone is explained by analysing radical distribution and rate constant as a function of temperature. At high equivalence ratio, the radical transportation and concentration are significantly increased. The high radical fractions in regions with over-adiabatic temperature contribute to the formation of hot spots directly under ultra-lean conditions. As the rate constants of reactions $\text{H} + \text{O}_2 (+\text{M}) = \text{HO}_2 (+\text{M})$ and $\text{HO}_2 + \text{OH} = \text{H}_2\text{O} + \text{O}_2$ decay exponentially with increasing temperature, their heat release rates are relatively enhanced in moderate-temperature regions, which contributes to a broadened reaction zone for the case with higher equivalence ratio. In summary, due to the combined effects of radical fractions and reaction rate constants, the local heat release changes in different temperature windows with varying mixture equivalence ratios.
8.1.4 Effects of integral length scale

Five high-Ka cases with different ratios of integral length scale to laminar flame thickness \((l/\delta_L)\) are performed under 1 atm and 5 atm to investigate the effects of the integral length scale and pressure on turbulent flame structures and chemical reactions. Statistics of mean curvature, tangential strain rate, flame thickness, flame area and heat release rate are examined to identify the effects.

For turbulent flames under the same Ka and pressure but varying \(l/\delta_L\) ratios, the flame fronts demonstrate different structures. At \(l/\delta_L = 0.5\), the reaction zone is continuous, and the heat release rate is comparable to that of the corresponding laminar flame. When the \(l/\delta_L\) ratio is increased higher than 1.0, the flame fronts are severely stretched and the heat release rate is dramatically enhanced. The convex region and concave region are characterised by high and low SDF values, respectively. With increasing \(l/\delta_L\) ratio, the turbulence effect on thickening flame becomes more apparent. However, statistical analysis shows that the PDF distributions of curvature and tangential strain rate are comparable between flames with the same Ka, even though the mean values are smaller at \(l/\delta_L = 0.5\). These findings indicate that when \(l/\delta_L < 1.0\), turbulence is unable to stretch and interrupt the reaction zone despite the high Ka, curvature and \(a_T\).

The turbulent flame with \(l/\delta_L < 1.0\) under the atmospheric pressure also exhibits laminar flame structures, which confirms that only large integral scales could modify flame structures. In the high Ka flame with unity \(l/\delta_L\) under the atmospheric pressure, the flame thickening effect increases from 400 to 1000 K and then starts to decrease until the thickening effect disappears. When pressure is elevated to 5 atm and the \(l/\delta_L\) ratio is kept at unity, the thickening effect increases dramatically with temperature. The
difference between the two flames under different pressures is attributed to the shrunken reaction layer toward high-temperature regions. It is interesting to note that the two flames present identical PDFs of scaled curvature and tangential strain rate, which suggests that keeping constant $l_\delta/\delta_L$ ratio and Ka can isolate the effects of pressure on flame front structures. Moreover, it is found that pressure and the $l_\delta/\delta_L$ ratio co-determine the probability of positive curvatures on the flame front, and the effect of $l_\delta/\delta_L$ ratio is more significant in the studied cases.

Due to the increased flame stretch, heat release is enhanced in turbulent flames under elevated pressures and the heat release peak moves to regions with high values of the progress variable. On the high Ka flame front, the local regions with high tangential strain rate are also characterised by high heat release rate under 5 atm, whereas it is not evident at the atmospheric pressure. The tangential strain rate reaches peak values near zero curvature regions and it decreases as the absolute value of curvature increases. Moreover, heat release contributions of elementary reactions show that the global chemical pathways are insensitive to the $l_\delta/\delta_L$ ratio but sensitive to pressure. In summary, the turbulent flame with unity $l_\delta/\delta_L$ ratio could capture the main features of heat release as those in flames with higher $l_\delta/\delta_L$ ratios. Pressure, on the other hand, could modify the chemical pathways.

8.2 Future work

The present work has performed direct numerical simulations of lean premixed H$_2$/air flames over a wide range of the Karlovitz number, pressure, equivalence ratio and integral length scale. The major effects of these parameters on flame structures and
chemical pathways have been addressed. It would be interesting to do further data
ing and extend this study to complex fuel combustion.

- As indicated in Chapter 5, the Peters’s regime diagram should be modified for
  high-pressure combustion. Meanwhile, the viscosity and fuel type should be
  considered. It is pointed out in Ref. [108] that the reaction zone Karlovitz
  number is better to characterise the flame structures than the classical Karlovitz
  number. Carlsson et al. [39] also proposed a species-specific Karlovitz number
  to support the analyse of flames with strong turbulence/chemistry interaction. It
  will be meaningful to develop a regime diagram which can accurately predict
  turbulent flame characteristics over a wide range of pressure.

- In turbulent flames, the flame front structure is determined by both strain and
  curvature. The present study has addressed the effects of curvatures. The effects
  of strain rate should also be studied. It will be interesting to clarify these effects
  on flame topology, especially at high Ka and elevated pressures.

- Syngas is a major product of biomass gasification, which can achieve carbon
  neutral in electricity generation. However, many fundamental issues related to
  syngas combustion at high Ka and under elevated pressures have not been
  clarified, which seriously affects the design of clean and efficient gas turbines.
  It would be meaningful to extend the current study to syngas combustion. In
  addition, the effects of H₂, CO and CH₄ on flame stabilization could also be
  investigated.
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Appendix A

Effects of Pressure on Turbulent H₂/air Flames in the Thin Reaction Zone

Three DNS cases of lean premixed turbulent H₂/air flames in the thin reaction zones are performed under 1, 2 and 5 atm respectively (presented in paper 1). These cases have the same equivalence ratio $\varphi = 0.6$ and the unburned gas temperature $T_u = 298$ K. Table 1 lists the detailed parameters of the three cases.

Table 1 Simulation parameters.

<table>
<thead>
<tr>
<th>Case</th>
<th>A</th>
<th>B</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pressure (atm)</td>
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<td>2</td>
<td>5</td>
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<tr>
<td>$S_L$(cm/s)</td>
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<td>47.9</td>
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<td>$\delta_L$(cm)</td>
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<td>657</td>
<td>657</td>
</tr>
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<td>305</td>
<td>762</td>
</tr>
<tr>
<td>Ka</td>
<td>77</td>
<td>62</td>
<td>64</td>
</tr>
</tbody>
</table>
Figure 1 shows the cumulative distribution function (CDF) of mean curvatures of flame front. With increasing pressure, the CDF decreases from 0.465 to 0.365, which means the convex regions are enlarged at elevated pressures.

![Figure 1](image)

Fig. 1 Cumulative distribution function of mean curvatures of flame front.
Appendix B

List of Publications

Journal articles


Conference proceedings


