Numerical modeling of thermal stratification driven combustion regimes

Ali Shahanaghi
Numerical modeling of thermal stratification driven combustion regimes

Ali Shahanaghi

A doctoral thesis completed for the degree of Doctor of Science (Technology) to be defended, with the permission of the Aalto University School of Engineering, at remote examination held at https://aalto.zoom.us/j/67561083932 on 8th of March 2024 at 12:00.

Aalto University
School of Engineering
Department of Mechanical Engineering
Energy Conversion and Systems Research Group
Supervising professor
Professor Ville Vuorinen, Aalto University, Finland

Thesis advisors
Professor Ossi Kaario, Aalto University, Finland
Doctor Shervin Karimkashi Arani, Aalto University, Finland

Preliminary examiners
Professor Matei Radulescu, University of Ottawa, Canada
Professor Huangwei Zhang, National University of Singapore, Singapore

Opponent
Professor Michael A. Liberman, Nordic Institute for Theoretical Physics, Sweden

Aalto University publication series
DOCTORAL THESES 40/2024

© 2024 Ali Shahanaghi

ISSN 1799-4934 (printed)
ISSN 1799-4942 (pdf)

Unigrafia Oy
Helsinki 2024

Finland
Abstract

This dissertation is related to the research areas of computational physics and chemistry. The purpose of the study is to determine the role of temperature stratification in the secondary combustion of gasoline surrogates within the end-gas of spark ignition (SI) engines. Specifically, it highlights the influence of the Negative Temperature Coefficient (NTC) on the prevalence of various combustion regimes, including spontaneous, subsonic, and supersonic ignition, detonation, and deflagration (flame), within one and two-dimensional (1D/2D) scenarios. Gaining a deeper understanding of these phenomena facilitates understanding of the interaction between pressure oscillations, reaction front propagation, and fuel chemistry at SI engine-relevant conditions. Computational fluid dynamics (CFD) simulations of reacting flows, incorporating finite rate chemistry, efficient simulation methods, and high-performance computing, enable the investigation of the underlying physical and chemical processes in such reacting flows.

This dissertation consists of three appended journal publications and their brief summary. In Publication I, a diagnostic approach distinguishes between secondary spontaneous ignition and flame of primary reference fuel (PRF) and PRF-ethanol (PRF-E) blends, in 1D setups. In Publication II, established 1D regime diagrams (detonation peninsula) are reconstructed for the PRF and PRF-E mixtures analyzing induced pressure levels from hotspot ignition (i.e. subsonic/supersonic ignition, and detonation modes). In Publication III, 2D phenomena are explored involving converging shock/detonation fronts and shockwave reflection-induced detonation, using the novel ARCFoam numerical framework. In essence, Publication I explores the prevalence of combustion modes at two extremes within the range of studied modes, namely flame and spontaneous ignition in a 1D setting. Publication II focuses on the pressure oscillations induced by hotspot ignition regimes within the 1D framework. Lastly, in Publication III, further geometrical considerations are introduced by investigation of hotspot ignition in 2D scenarios.

The main conclusions of this dissertation are as follows: 1) NTC chemistry of the studied fuels is the dominant factor in manipulating the reactivity, inducing secondary combustion modes, and amplified pressure levels. 2) Given the dominance of auto-ignition in the presence of NTC and the impact of thermal stratification on ignition characteristics, density-based 1D/2D CFD simulations are preferred for analyzing underlying combustion regimes. 3) While the detonation peninsula predicts the initial ignition regime induced by hotspot ignition, NTC chemistry and 2D effects such as front curvature and shock reflection can significantly alter combustion dynamics. 4) Apart from the complexity of the flow field and the employed numerical techniques, the resource-intensive nature of finite rate chemistry solution remains the primary bottleneck in such reactive CFD simulations.

Keywords Combustion Regimes, Thermal Stratification, Spark Ignition Engines, Knock, Finite Rate Chemistry, Primary Reference Fuel

<table>
<thead>
<tr>
<th>ISBN (printed)</th>
<th>ISBN (pdf)</th>
<th>ISSN (printed)</th>
<th>ISSN (pdf)</th>
</tr>
</thead>
<tbody>
<tr>
<td>978-952-64-1688-5</td>
<td>978-952-64-1689-2</td>
<td>1799-4934</td>
<td>1799-4942</td>
</tr>
</tbody>
</table>

Location of publisher Helsinki | Location of printing Helsinki | Year 2024

Preface

This dissertation was conducted during the years 2019-2023, at Aalto University School of Engineering, in the Energy Conversion research group. This research has been financially supported by Neste corporation with the Digifuels project and the Mechanical Engineering Dean’s funding.

First and foremost, I would like to thank Professor Matei Radulescu (University of Ottawa) and Professor Huangwei Zhang (National University of Singapore) for their time and efforts as pre-examiners of my dissertation. I am grateful for their positive and constructive feedback. I also thank Professor Michael A. Liberman (Nordic Institute for Theoretical Physics), once again, for acting as the opponent in the public defense.

I extend my gratitude to my supervisor, Professor Ville Vuorinen. His remarkable ability to grasp complex issues, consistent availability to address my queries, and invaluable assistance in overcoming challenges beyond the realm of research have left an indelible mark on my journey. I am equally indebted to my thesis advisor and project manager, Professor Ossi Kaario. His trust, support, and guidance have been the bedrock upon which this dissertation stands. His patience and attention to my research during the hectic times of the pandemic were instrumental in enabling the completion of this thesis.

A special acknowledgment goes to Dr. Shervin Karimkashi Arani, my advisor and trusted friend, with whom I dedicated countless hours to engaging in discussions and planning to overcome obstacles in simulations, publications, and research endeavors. His collaboration resembled an additional processing power throughout these four years. I express my gratitude to Professor Marti Larmi for entrusting me with being a lecturer in the Thermochemical Energy Conversion course from 2020 to 2023, during which I gained invaluable insights. I'd like to extend my appreciation to the DigiFuels team from Neste and Aalto University for their encouragement and continued support of my research.

My sincere thanks go to Dr. Hadi Pasdarshahri, my Master’s thesis supervisor, who mentored me in becoming a researcher. I will forever be grateful for his kindness and guidance during my time at Tarbiat Modares.
University. To Professors K. Mazaheri, H. Khaleghi, R. Madahian, and M. Zabetian, who introduced me to the captivating intricacies of combustion, computational fluid dynamics, multiphase flows, and internal combustion engines, I am profoundly thankful. Their selfless and thoughtful teaching opened the door to the profound world of complex yet harmonious computational physics and chemistry and its applications.

I would like to express my gratitude to my senior colleagues, Heikki, Petteri, Erkki, and Atma, for their generous instructions when I began my PhD journey. Throughout these years, I have drawn inspiration from their work commitment and transparent approach to their tasks. I also extend my thanks to the members of our CFD group and my colleagues at Aalto for their positivity and professionalism, which greatly contributed to my own work. I’d like to acknowledge Mahmoud, Jeeva, Vili-Petteri, Stas, Gizem, Alpo, Parsa, Ali Ashnani, Bayazid, Tolga, Ilya, Nushrat, Jennifer, Aleksi, Zayn, and Ali Haider for their friendship and support. I will always cherish the fond memories we’ve created together and will continue to root for your success. Lastly, I want to offer my heartfelt appreciation to my dearest friends, Mehdi, Parham, and Shahrooz. Their help was indispensable during the challenging moments throughout these years.

Last but certainly not least, I want to dedicate this work to my parents, whose steadfast support and countless sacrifices have made my dreams a reality. To my brother Alireza, his unwavering support has been a pillar of strength. And to my father, teacher, and motivator, Prof. Kamran Shahanaghi, his support throughout both the highs and lows of my journey means the world to me. I hope to have made him proud.

Espoo, Finland, February 11, 2024,

Ali Shahanaghi
# Contents

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>Preface</td>
<td>1</td>
</tr>
<tr>
<td>Contents</td>
<td>3</td>
</tr>
<tr>
<td>List of Publications</td>
<td>5</td>
</tr>
<tr>
<td>Author's Contribution</td>
<td>7</td>
</tr>
<tr>
<td>List of Figures</td>
<td>9</td>
</tr>
<tr>
<td>List of Tables</td>
<td>13</td>
</tr>
</tbody>
</table>

## 1. Introduction
- 1.1 Scope of thesis .......................... 15
- 1.2 Motivation ................................ 15
- 1.3 Engine relevant information ............... 17
- 1.4 Combustion modes .......................... 20
- 1.5 Deflagration versus ignition front ....... 21
  - 1.5.1 Front characteristics .................. 21
  - 1.5.2 Beta curve theory ....................... 22
- 1.6 Detonation versus Deflagration ............ 24
- 1.7 Detonation structure ....................... 27
  - 1.7.1 ZND structure of detonations .......... 28
  - 1.7.2 Two-dimensional structure of detonations .... 29
- 1.8 Hotspot Ignition Regimes ................... 32
- 1.9 Objectives of the dissertation ............. 34
- 1.10 Outline of the dissertation ............... 35

## 2. Methodology
- 2.1 Governing equations ......................... 37
  - 2.1.1 Pressure based solution ................. 38
  - 2.1.2 Density based solution ................. 39
- 2.2 Finite rate chemistry solution ............. 41
  - 2.2.1 Chemical source term evaluation ....... 42
# Contents

2.2.2 Integration of kinetic ODEs ......................................... 43
2.3 Simulation efficiency improvement ................................. 44
2.3.1 Adaptive Mesh Refinement ................................. 45
2.3.2 AMR mesh balancing ........................................ 46
2.3.3 Chemistry problem balancing ................................. 47

3. Summary of publications ............................................. 49
3.1 Publication I .................................................. 49
3.2 Publication II .................................................. 53
3.3 Publication III .................................................. 56

4. Conclusions and perspectives ......................................... 61
4.1 Conclusion ..................................................... 61
4.2 Limitations .................................................... 62
4.3 Future work ................................................... 63

A. Code snippets ..................................................... 67
A.1 Creating coarse cell maps ........................................ 67
A.2 AMR mesh balancing ........................................... 69
A.3 redecomposePar utility .......................................... 72

References ............................................................. 77

Publications ........................................................... 85
This thesis consists of an overview and of the following publications which are referred to in the text by their Roman numerals.


Author’s Contribution

Publication I: “A Diagnostic Approach to Assess the Effect of Temperature Stratification on the Combustion Modes of Gasoline Surrogates”

The author was the lead author and responsible for theoretical and numerical analysis. V. Vuorinen and S. Karimkashi supported in building the theoretical formalism of the beta curve theory. It should be noted that the paper draft has been improved by the comments provided by all co-authors.

Publication II: “Temperature Stratification Induced Ignition Regimes for Gasoline Surrogates at Engine-Relevant Conditions”

The author was the lead author and responsible for theoretical and numerical analysis. The presented density-based solver in this paper partially incorporates the solver previously developed by V. Vuorinen. It should be noted that the paper draft has been improved by the comments provided by all co-authors.

Publication III: “Efficient two-dimensional simulation of primary reference fuel ignition under engine-relevant thermal stratification”

The author was the lead author and responsible for theoretical and numerical analysis. The presented density-based solver in this paper partially incorporates the solver previously developed by V. Vuorinen. It should be noted that the paper draft has been improved by the comments provided by all co-authors.
List of Figures

1.1 Combustion parameters of engine knock cycle, demonstrating pressure trace, pressure oscillation, heat release rate (HRR) and unburned gas temperature (T). The figure is obtained from the work by Wang et al. [90] and reprinted with permission from Elsevier. .................................................. 18

1.2 Schematic presentation of combustion mode diagram, the solid line ($\beta = 1$) indicates the border between the spontaneous ignition and the deflagration modes. Theoretical $\beta$ values ($C_\beta = 1$) are also outlined (dashed lines). In practice, as noted also in Publication I, the transition region around the $\beta$-curve may manifest features from both combustion modes. .................................................. 24

1.3 Schematic presentation of combustion wave with fixed coordinates to the wave. .................................................. 25

1.4 Hugoniot curve corresponding to $q = q_1$, originating from point A at ($p_1, v_1$). Note that the curve does not precisely intersect the so-called origin. The black dashed lines passing through point A represent Rayleigh lines. The blue shade represents physically inaccessible solutions. .................. 27

1.5 ZND structure of H2/air mixture at pre-shock conditions of $P_0=6670$ pa and $T_0=300$ K, showcasing normalized profiles of pressure, temperature, and density. The figure highlights key features including the shock front, induction and exothermic lengths. The normalized CJ equilibrium values for pressure ($\frac{p}{p_{CJ}}$), temperature ($\frac{T}{T_{CJ}}$), and density ($\frac{\rho}{\rho_{CJ}}$) are indicated by horizontal dashed lines. The top axis presents the corresponding time frame, initiated from the shock front propagating with CJ speed. .................. 28
1.6 Two-dimensional detonation front structure, illustrating the triple point trajectory (detonation cell), Mach shock, and shear layers. The figure is obtained from the work by M. I. Radulescu [69] and reprinted with permission from Elsevier. .......................................................... 30

1.7 2D planar detonation of H$_2$/air at $P_0=6670$ pa and $T_0=300$ K, a) schematic presentation of the simulation setup, b) detonation cellular structures inside the channel, c) sensitivity of the triple point trajectories to the grid resolution. Image from Publication III. .......................... 31

1.8 Reproduced ignition regime diagrams from 1D simulations Publication II at $P = 50$ bar and $T_{ave} = 900$ K. Blue and pale blue colors in the top-right/bottom-left regions indicate sub/supersonic ignition, while the red region denotes the detonation regime. The black dashed line represents the original Bradley detonation peninsula. The purple and green dash-dotted lines indicate regions associated with off-centered ignition and coolspots, respectively. Points 1-3 correspond to the selected points for 2D cylindrical simulations, the image is from Publication III. ............... 34

2.1 A schematic presentation of the main processes taken within a time step in reactive compressible solution in ARCFoam, flow chart reconstructed from Publication III. .......................... 45

2.2 Schematic demonstration of a) visible cells in the two refinement regions, ($w_1, w_2$) and ($w'_1, w'_2$) indicate local weights (number of cells) of each refinement region, b) different cell levels (1 and 2) on each refinement region and their projection on parent cells (level 0), c) mesh and temperature distribution of Richtmyer–Meshkov instability in H$_2$ detonation with 5 levels of refinement, image from Publication III. .......................... 46

3.1 Schematic representation of the combustion process: step I - Thermal diffusion, step II - Primary ignition, step III-a - Spontaneous ignition mode with an ignition front propagating towards the unburnt mixture, and step III-b - Deflagration mode with a premixed flame propagating towards the unburnt mixture. .......................................................... 50

3.2 PRF and PRF-E ignition delay time versus temperature profiles at, a: $P = 20$ and b: 50 bar, black circles are marking the locations of the studied cases. .......................... 51
3.3 Combustion mode distribution of case IV (P = 50 bar and \( T_{ave} = 900 \) K), black and white regions represent spontaneous ignition and deflagration modes of a) PRF and b) PRF-E mixtures respectively. Colored lines display the \( \beta \)-curves calculated using different \( C_\beta \) values, Eq. 1.10, c) IDT distribution at the initial time versus T of PRF and PRF-E mixtures.

3.4 A schematic presentation of the combustion process shows, (a) hotspot/coolspot initial T profiles, (b) centered ignition and regular propagation inside the hotspot/coolspot, (c) off-centered primary ignition due to NTC and double ignition front propagation, and d) occurrence of primary ignition and subsequently, formation of the secondary ignition kernel due to NTC.

3.5 Ignition regime diagram of case I (P = 50 bar and \( T_{ave} = 900 \) K), red zone indicates the detonation peninsula of (a) PRF and (b) PRF-E mixtures, respectively. Colored circles display the hotspot pressure calculated for each 1D simulation. Letters P, B, and N indicate supersonic ignition, subsonic ignition, and the nominal condition for knock initiation inside SI engines [38]. (c) IDT distribution at the initial time versus T of PRF and PRF-E mixtures. The black dashed line represents the original Bradley detonation peninsula.

3.6 Two-dimensional contour plots illustrating the characteristics of H2 cylindrical detonation: a) Cellular structure showing the presence of a no-cell region (I) and a cell growing region (II). Density gradient (b) and temperature (c) profiles at different time instances: \( t_1 = 0.000155 \) s, \( t_2 = 0.00049 \) s, \( t_3 = 0.000825 \) s, \( t_4 = 0.001165 \) s. d), e), and f) Zoomed-in profiles depicting the OH mass fraction, pressure, and temperature at the specified location within the box.

3.7 Transient profiles of temperature and normalized pressure at point 2 for the PRF mixture at \( T = 900 \) K and \( P = 50 \) bar, demonstrating the appearance of off-centered ignition and subsequent converging/diverging detonation.
List of Tables

1.1 RON and MON tests operating conditions . . . . . . . . . . 19
1. Introduction

1.1 Scope of thesis

The present dissertation contributes to the field of computational physics and chemistry. The incorporated research method integrates computational fluid dynamics with reactive flow modeling and computational chemistry approaches. This work investigates the influence of temperature stratification on combustion dynamics in the presence of low-temperature chemistry of gasoline surrogates, particularly the negative temperature coefficient, under scenarios resembling knocking in spark ignition engines. In Publication I, the study explores the prevalence of deflagration versus auto-ignition in a one-dimensional setting. Building upon this, in Publication II we investigate scenarios characterized by high-speed combustion and the emergence of shock waves. Finally, Publication III presents the two-dimensional characteristics of these phenomena by introducing an efficient computational approach. Collectively, these publications provide a comprehensive understanding of end-gas combustion dynamics, spanning from one-dimensional analyses to more complex scenarios involving shock waves and geometrical effects.

1.2 Motivation

Despite the emergence of alternative technologies such as battery electric vehicles, biofuels, and hydrogen, substantial barriers impede their rapid adoption. These alternatives necessitate comprehensive life-cycle analyses, and imposing them prematurely could result in adverse environmental, economic, and social consequences [31, 79]. As a result, global transportation continues to be dominated by internal combustion engines (ICEs) and liquid fuels derived from petroleum. By 2040, conventional combustion engines are projected to supply around 85-90% of transport en-
Energy [46]. Enhancing the efficiency and emissions performance of internal combustion engines (ICEs) becomes crucial to meet emission goals. Practical approaches already in the market show promise, with leading spark ignition (SI) engines in the U.S. exhibiting 14% lower fuel consumption than average [46]. Advancements in engine and powertrain technologies can reduce light-duty vehicle fuel consumption by over 30%, and further reductions to 50% are feasible with hybridization and light-weighting [46]. Despite plans to phase out ICE vehicles, investing in IC engine research remains important for substantial improvement potential. In sectors like heavy-duty transport where electric solutions face challenges, IC engine research and renewable e-fuels offer emission reduction possibilities [31, 46].

Given the dominance of gasoline-fueled SI engine vehicles in the 2020 market [11] and their anticipated continued prevalence, enhancing SI engine efficiency becomes essential to achieve CO\textsubscript{2} targets. The efficiency of SI engines relies on factors such as the compression ratio, with maximizing this ratio being crucial to minimize CO\textsubscript{2} emissions. However, this optimization is strongly constrained by the knocking phenomenon.

The fundamental knowledge on knock is well-established [11, 90, 32], yet, comprehending the correlation between knock and factors like fuel chemistry, pressure oscillations, heat transfer, and auto-ignition front propagation is still in its early stages [75, 90]. Octane numbers (ONs), which encompass RON and MON (research and motor octane numbers) [2, 3] tests, have been devised to evaluate fuel performance. These tests measure pressure intensity in a cooperative fuel research (CFR) engine at a specific compression ratio, mimicking the behavior of primary reference fuel blends containing iso-octane and n-heptane. However, limitations persist in this approach. The influence of CFR engine conditions and fuel composition on ONs raises questions about ONs compatibility with various engine technologies or for high-knock-resistance fuels (exceeding ONs=100) [8]. Notably, reported ON values may not reveal the complex process underlying specific pressure intensities, thereby not fully explaining the role of fuel thermophysical properties and the engine operational conditions on the knocking process.

To resolve these uncertainties, this dissertation centers on the thermophysical processes giving rise to abnormal combustion under conditions relevant to SI engines. The objective is to attain a better comprehension of combustion dynamics and characteristics of gasoline surrogates under varying temperature/pressure conditions. By exploring the underlying thermophysical mechanisms, we can better elucidate the complexities of combustion and pressure wave generation processes in modern engine technologies.
1.3 Engine relevant information

The occurrence of knock severely limits SI engine efficiency as it involves spontaneous combustion of the air-fuel mixture ahead of the flame front. Knocking phenomena can be classified as mild, strong, or superknock based on the amplitude of pressure oscillations. Mild and strong knock may result from localized hot spots causing pressure fluctuations in the order of $\Delta P \approx 5$-24 bar. Superknock, triggered by pre-ignition, is an extreme event with pressure fluctuations in the order of $\Delta P \gtrapprox 50$ bar, often accompanied by detonation. Superknock may arise due to shock wave interaction with a reaction front on surfaces or within a hotspot. All forms of knock are commonly considered to have a negative impact on engine performance, emissions, and durability [32, 91, 36, 75].

In SI engines, the combustion process involves various stages including the premixing of fuel and air, followed by compression, and energy release through an expanding turbulent flame ignited by the spark. As combustion progresses, the temperature and pressure rise within the unburnt mixture ahead of the flame front, known as the end-gas [32]. The end-gas is not completely homogeneous, characterized by temperature stratification exceeding 20 K [90], mainly due to the turbulent mixing of hot gases within the cylinder with the fresh charge and existing hotspots [38]. Autoignition, which arises from one or more hotspots, is triggered based on the pressure/temperature history and the fuel anti-knock properties. The likelihood of auto-ignition and subsequently knocking increases when more time is available for chemical reactions in the end-gas, such as in low engine speeds or larger engines. The chemical energy stored in the end-gas from the unburnt fuel can be rapidly released through auto-ignition, leading to a rapid pressure rise and the formation of pressure waves within the cylinder. This process is schematically illustrated in the lower panel in Figure 1.1.
Combustion parameters of engine knock cycle, demonstrating pressure trace, pressure oscillation, heat release rate (HRR) and unburned gas temperature (T). The figure is obtained from the work by Wang et al. [90] and reprinted with permission from Elsevier.

The pressure trace, pressure oscillation, heat release rate (HRR), and unburned gas temperature (T) of a typical knocking case are demonstrated in Fig. 1.1. The upper panel demonstrates the pressure measured inside the cylinder versus the crank angle degree (CAD) and the associated oscillations resulted from high-pass filtering the pressure signal. HRR \([\text{MJ}/(\text{m}^3.\text{deg})]\) is computed in accordance with the first law of thermodynamics [90], as follows:

\[
\text{HRR} = \left(\frac{\gamma}{\gamma - 1}\right) P \frac{dV}{d\theta} + \left(\frac{1}{\gamma - 1}\right) V \frac{dP}{d\theta}, \tag{1.1}
\]

where \(\gamma\) is the specific heat ratio. In the calculation of unburnt temperature, an adiabatic end-gas compression process is assumed,

\[
\int_{T_0}^{T(t)} \frac{\gamma}{\gamma - 1} d(lnT) = ln \frac{p(t)}{p_0}. \tag{1.2}
\]

The RON and MON have been adopted for gasoline fuels [27] to assess the knock resistance of fuels in SI engines. Fuels with higher octane numbers suggest more resistance to auto-ignition. The RON and MON tests have standard procedures in the CFR engine [56]. Primary Reference Fuel (PRF) which are binary mixtures of \(n\)-heptane and iso-octane are used as references for measuring the knock intensity (KI) of the test fuels. KI is quantified by the maximum peak-to-peak amplitude of the knock.
signal, obtained by filtering the in-cylinder pressure signal to eliminate high- and low-frequency components. The operating conditions for the RON and MON tests [2, 3], as shown in Table 1.1, are different; MON is tested in a more severe situation, where the intake temperature is higher and the engine speed is higher.

Table 1.1. RON and MON tests operating conditions

<table>
<thead>
<tr>
<th>Operating condition</th>
<th>RON</th>
<th>MON</th>
</tr>
</thead>
<tbody>
<tr>
<td>Intake air temperature</td>
<td>52°C</td>
<td>149°C</td>
</tr>
<tr>
<td>Intake air pressure</td>
<td>1 atm</td>
<td>1 atm</td>
</tr>
<tr>
<td>Coolant temperature</td>
<td>100°C</td>
<td>100°C</td>
</tr>
<tr>
<td>Engine speed</td>
<td>600 RPM</td>
<td>900 RPM</td>
</tr>
<tr>
<td>Spark timing</td>
<td>13°bTDC</td>
<td>14-26°BTDC</td>
</tr>
<tr>
<td>Compression ratio</td>
<td>4-18</td>
<td>4-18</td>
</tr>
</tbody>
</table>

For non-PRF fuels, RON can be higher than MON and the sensitivity (S) of the fuel is defined as $S = RON - MON$. Since engine operating conditions vary significantly, RON and MON are designed as limiting operating conditions but are not enough to describe the knock resistance of all fuels. For instance, ethanol addition to gasoline (biogasoline) can enhance the knock resistance of the fuel mixture boosting RON to beyond 100 [8]. The addition of ethanol mitigates the negative temperature coefficient (NTC) behavior of the mixture which may increase the ignitability of the mixture at intermediate temperatures. However, the presence of ethanol increases the resistance to ignition in low temperatures where knock typically occurs. Finally, at high temperatures, the fuel mixture obtains ignition characteristics similar to gasoline [16]. To take into account the whole range of engine operating conditions, Kalghatgi et al. [33, 34] proposed the Octane Index (OI), defined as,

$$OI = RON - K \times S.$$  

(1.3)

The K factor depends on the engine operating conditions and is supposed to be independent of fuel properties. Notably, for the most knock-limited regions in modern SI engines [72, 35] the value of K tends to become negative. Advancements in engine design, including improvements in cooling systems, boost systems, and injection systems, have resulted in reducing the value of parameter K [57]. Consequently, the operational conditions of modern engines often surpass the boundaries outlined by the RON and MON tests, which were originally designed for engines equipped with technologies similar to the CFR engine, i.e. utilizing heated carburetors and featuring insufficient cooling systems [57]. In modern SI
engines, unburnt gas temperature (e.g. at a compression pressure of 15 bar, Tcomp_{15} [37]) tends to be lower due to the lower intake temperature than that in the RON and MON tests. Although the fuel ignition indices, RON, MON, and OI, have been used to describe fuel anti-knock properties in automotive engineering, the relationship between RON/MON and OI/K is still not clear. This is due to the fact that auto-ignition in internal combustion engines is directly associated with the combustion chemistry of hydrocarbons at different operating conditions. Therefore, a further understanding of underlying physics and chemistry is necessary to more accurately characterize knocking phenomena.

### 1.4 Combustion modes

Reaction fronts can be defined as narrow regions characterized by high heat production from rapid reactions [45]. Combustion modes, on the other hand, encompass the spatial and temporal characteristics of these reaction fronts. In Publication I - Publication III, we explore and characterize the combustion modes of PRF87 (87% iso-octane/13% n-heptane by volume, referred to as PRF hereafter) and PRF-ethanol (18% n-heptane/62% iso-octane/20% ethanol by volume, referred to as PRF-E hereafter) mixtures with air.

The classification of reaction fronts is based on their attributes, such as propagation speed, as well as their characteristics, including reaction rates and key thermophysical events involved in energy transport. For instance, in SI engines, abnormal combustion mode is associated with a type of combustion that leads to excessive pressure rise in the cylinder through rapid energy release. However, the abnormal combustion mode can further be categorized based on the intensity of generated pressure fluctuations, adding complexity to the understanding of combustion dynamics.

In the following sections, we analyze the distinction between deflagration and ignition front, introducing the concept of the beta curve theory to explain their relative dominance. Subsequently, we compare deflagration and detonation processes, exploring relevant theories that shed light on their differences and underlying mechanisms. Understanding these combustion modes along with the background theories is crucial for comprehending the complex phenomena occurring during combustion processes leading to knocking and can significantly impact the design and performance of combustion engines.
1.5 Deflagration versus ignition front

In Publication I, we examined the prevalence of secondary combustion within the end-gas of SI engines, specifically deflagration and spontaneous ignition modes. The following sections introduce the distinctive characteristics of deflagrations and ignition fronts and present a diagnostic theory to distinguish between deflagrations and spontaneous ignition.

1.5.1 Front characteristics

In the absence of turbulence, a laminar deflagration (flame) initiates from a weak ignition source within a combustible mixture [9]. The propagation of the flame into the reactants ahead occurs through the diffusion of heat and mass. The steep gradients in temperature and chemical species concentration across the flame front facilitate the transport of heat and radical species, leading to ignition in the reactants ahead [48]. As a result, a deflagration front resembles a diffusion wave, and its velocity is proportional to the square root of the diffusivity and reaction rate [67],

\[ S_l \propto \sqrt{D\Omega} \] (1.4)

where \( D \) and \( \Omega \) represent the diffusivity and reaction rates, respectively. Consequently, flame propagation can be categorized into two types based on these controlling parameters: diffusion-controlled (governed by \( D \)) and chemically controlled (governed by \( \Omega \)). The controlling parameter determines the slower process that dictates the flame speed (\( S_l \)). However, diffusion of heat and active radicals typically remains a definitive parameter influencing the laminar flame speed. Premixed flame structure is commonly divided into two regions: the preheat zone and the reaction zone [88]. The significance of the preheat zone lies in increasing the temperature for follow-up reactions and producing radical pools. In addition, the preheat zone plays a crucial role in the dynamics of the reaction front propagation when the flame front experiences curvature or scenarios involving non-unity effective Lewis numbers [9].

On the other hand, ignition front propagation is solely dependent on the sequential auto-ignition of the reacting mixture in time and space where diffusion effects are negligible [94]. Hence in such scenarios, the favorable combination of heat release and reactivity gradient controls the propagation speed. Zeldovich described the ignition of a nonuniform mixture in several distinct regimes of propagation, including deflagration and ignition front propagation. He further demonstrated that the ignition front speed (\( S_{ign} \)) is inversely proportional to the initial temperature gradient. Assuming a homogeneous mixture, the ignition delay time (IDT) is solely a function of temperature fluctuation and can be written as:
\[ S_{\text{ign}} = \frac{1}{|\nabla \tau|} = \left(\frac{\partial \tau}{\partial T} \right)^{-1}, \]  

where \( \tau \) represents IDT. The above equation suggests that when the temperature gradient is low, \( S_{\text{ign}} \) converges towards very high values resembling a volumetric spontaneous ignition (thermal explosion), and when it is high the \( S_{\text{ign}} \) vanishes and the reaction front stabilizes the molecular transport process as deflagration. Based on such a notion Sankaran et. al. [77] proposed a criterion to distinguish between the two regimes by utilizing their propagation speeds,

\[ \beta = C_\beta \frac{S_l}{S_{\text{ign}}}, \]

where \( C_\beta \) is a coefficient in the order of unity. The theory suggests that if \( \beta \ll 1 \), the reaction front propagation is driven by subsequent auto-ignition of the mixture as a result of the low gradient in IDT distribution. However, the condition \( \beta > 1 \) implies that the low reactivity of the unburnt mixture adjacent to the reaction front will allow the deflagration front development within the mixture. Notably, scenarios with \( \beta \approx 1 \) indicate that the reaction zone may display characteristics from both combustion modes.

### 1.5.2 Beta curve theory

In Publication I, the authors assessed the validity of the \( \beta \) criterion in terms of predicting secondary combustion events in the end-gas of SI engines. As discussed in the earlier sections, normal/mild knock is typically attributed to isolated pressure oscillations (\( \Delta P \approx 5-24 \) bar) caused by the auto-ignition of the mixture in the end-gas. Therefore, the \( \beta \) criterion was employed to categorize the secondary combustion mode between spontaneous ignition and deflagration under various temperature stratification levels. The study aimed to determine the length scales (\( \lambda \)) and amplitudes (\( \delta \)) of temperature variation at which spontaneous ignition and deflagration were dominant under low Mach number assumption (Mach < 0.3) and subsequently explore the influence of the fuel knock index on these combustion modes. To conduct a systematic study, the authors adapted and extended the theoretical/numerical investigation originally proposed by Karimkashi et al. [40] for mixture stratification in compression ignition engines, referred to as the \( \beta \)-curve diagnosis.

The extended \( \beta \)-curve diagnosis assumes an initial sinusoidal temperature stratification at specified average temperatures (\( T_{\text{ave}} \)), \( T(x = 0, t) = \delta \sin(kx) \), where \( k = \frac{2\pi}{\lambda} \) is wave number, with periodic boundaries \( T(x = 0, t) = T(x = \lambda, t) \). Subsequently, 1D heat diffusion equation can be written as:

\[ \frac{\partial T}{\partial t} = \frac{\nu}{\partial x^2}. \]
Here, $\nu$ is dynamic viscosity, and the equation assumes a constant heat diffusivity and unity Prandtl number. By solving Eq. 1.7 with the specified initial and boundary conditions (the details of which are provided in Publication I), we can determine the maximum temperature gradient in the domain, expressed as:

$$\left(\frac{\partial T}{\partial x}\right)_{t=\tau}^{\text{max}} = \delta k \exp(-k^2 \nu \tau).$$  \hfill (1.8)

After substituting Eq. 1.8 into Eq. 1.5 the ignition front speed at the instant of ignition is obtained,

$$S_{\text{ign}}|_{t=\tau} = \left(\alpha \delta k \exp(-k^2 \nu \tau)\right)^{-1}. \hfill (1.9)$$

Here $\alpha = \frac{\partial \tau}{\partial T}$ at $T_{ave}$ is an estimation for local ignition delay sensitivity to temperature variations. Further, substituting Eq. 1.9 into Eq. 1.6 gives

$$\beta = C_\beta S_{\text{fl}} \alpha \delta k \exp(-k^2 \nu \tau). \hfill (1.10)$$

Finally, considering $k = 2\pi/\lambda$, the relation between $\lambda$ and $\delta$ can now be written as:

$$\delta = \frac{\beta \lambda \exp(4\pi^2 \nu \tau \lambda^2)}{C_\beta S_{\text{fl}} \alpha 2\pi}. \hfill (1.11)$$

Equation 1.10 represents the modified $\beta$-curve equation specifically tailored for thermal stratification. By inserting $\beta = 1$ into Eq. 1.11, we obtain the borderline between the deflagrative and spontaneous ignition modes in the $(\delta, \lambda)$ space. The studied length scales are normalized by the diffusion length scale defined as $\lambda_{\text{diff}} = 2\pi \sqrt{\nu \tau}$. A schematic plot of the $\beta$-curve is depicted in Fig. 1.2, where the solid line ($\beta = 1$) theoretically separates the spontaneous ignition and the deflagration modes. However, in practice the borderline definition is not precise and the relative importance of the two modes is influenced by the temperature sensitivity of the induction kinetics, denoted as $\alpha$. For further details, please refer to Publication I. Different theoretical $\beta$ values ($C_\beta = 1$) are also shown as dashed lines. Different $\beta$ values indicate how the ignition front speed compares to the laminar flame speed, with $\beta > 1$ ($\beta < 1$) signifying the prevalence of the deflagration (spontaneous ignition) mode.
In the given notation, $\tau$, $S_l$, and $\alpha$ are all dependent on $T_{ave}$, pressure, and fuel composition. The ignition delay time ($\tau$) and laminar flame speed ($S_l$) can be estimated using the 0D constant volume reactor and 1D steady premixed flame models, respectively. It is important to mention that the choice of $\lambda/\lambda_{diff}$ being less than 2 in Fig. 1.2 is based on physical considerations to avoid strong ignition and subsequent pressure oscillations. A summary of the findings of Publication I is provided in the section 3.1.

1.6 Detonation versus Deflagration

According to the classical definition, [88, 48], deflagration refers to a reaction front that propagates at subsonic velocities, while detonation is characterized by supersonic propagation speeds. However, as discussed in the previous section, another attribute completing this categorization is the driving force behind the front propagation. Deflagration fronts are primarily driven by the diffusion process involving heat and mass transfer, whereas detonation fronts are derived by leading shock compressions. Consequently, the conditions that facilitate these processes become crucial, such as mixing and molecular diffusivities in deflagrations, and gas-dynamic coupling and initiation energy in detonations. In Publication II and Publication III, the authors investigated the different hotspot induced ignition regimes including detonation propagation in 1D and 2D scenarios. Therefore, in the following, the gas-dynamic theory of
one-dimensional steadily propagating combustion waves is discussed, and more information regarding steady/unsteady detonations is provided in later sections.

The schematic demonstration of the combustion wave and corresponding flow properties with coordinates relative to the front is presented in Fig. 1.3. The basic conservation equations include mass, momentum, and energy across a combustion wave with the coordinate system fixed to the wave are:

\[
\begin{align*}
\rho_0 u_0 &= \rho_1 u_1, \quad (1.12) \\
p_0 + \rho_0 u_0^2 &= p_1 + \rho_1 u_1^2, \quad (1.13)
\end{align*}
\]

and

\[
h_0 + \frac{u_0^2}{2} = h_1 + \frac{u_1^2}{2}. \quad (1.14)
\]

In the above equations, the subscripts 0 and 1 indicate reactants and products, respectively. \(h = h_f + \int c_p dT\), \(\rho\), \(p\), and \(u\) are enthalpy, density, pressure, and velocity, respectively, and \(h_f\) shows the enthalpy of formation. We may separate the enthalpy of formation and write the Eq. 1.14 in sensible enthalpy form as

\[
h_{s0} + q + \frac{u_0^2}{2} = h_{s1} + \frac{u_1^2}{2}, \quad (1.15)
\]

where \(q\) is the difference between the enthalpies of formation of reactants and products. Solving Eqs. 1.12 and 1.13 and using specific volume, \(\nu = 1/\rho\) we have:

\[
\frac{p_1 - p_0}{\nu_0 - \nu_1} = \rho_0^2 u_0^2 = \rho_1^2 u_1^2 = m^2, \quad (1.16)
\]

where \(m^2\) is flow rate. Using Eq. 1.16 and fixing the reactant state \((p_0, \nu_0)\) and flow rate results in the Rayleigh line. Increasing/decreasing flow rate results in increasing/decreasing the slope of the Rayleigh line pivoting through the reactant state point \((p_0, \nu_0)\), sweeping area (quadrants with no shade in Fig. 1.4) in the p-\(\nu\) space. In the limit of infinite/zero flow.
rate, the line becomes vertical/horizontal and the area swept indicates physically possible product states \((p_1, \nu_1)\). Finding the velocities \(u_0, u_1\) from the equation 1.16, e.g.

\[
u_0^2 = \frac{1}{\rho_0^2} \frac{p_1 - p_0}{v_0 - v_1},
\]

and substituting in energy equation 1.15 gives

\[
h_{s1} - (h_{s0} + q) = \frac{1}{2}(p_1 + p_0)(v_0 + v_1).
\]

The equation above represents the Hugoniot curve, which characterizes the set of product states corresponding to a given reactant state. Notably, when \(q\) is zero, Eq. 1.18 represents the Hugoniot curve for a non-reacting shock wave. By assuming an ideal gas equation of state and eliminating the sensible enthalpy \(h_s = \gamma \rho \frac{p}{\gamma - 1}\), the Hugoniot curve equation can be simplified to \(f(p, \nu) = 0\).

Figure 1.4 illustrates the Hugoniot curve for a known \(q = q_1\) and the reactant state at point A\((p_0, \nu_0)\). The blue-shaded quadrants in Fig. 1.4 represent physically inaccessible solutions, as they violate the flow rate constraints \(0 \leq \dot{m} \leq \infty\), while the quadrants with no shade contain all possible flow rates. The Rayleigh line and Hugoniot curve exhibit two tangency solutions: the minimum-detonation-velocity solution, known as the Chapman–Jouguet (CJ) detonation, and the maximum-deflagration-velocity solution, referred to as the CJ deflagration (Fig. 1.4). The tangency condition plays a crucial role in closing the conservation laws. However, the physical validity of these solutions relies on their agreement with experimental observations or other physical considerations beyond the conservation laws.
Introduction

At the CJ detonation point the detonation velocity (i.e., slope of the Rayleigh line) is minimum (CJ speed). Above this minimum velocity, the Rayleigh line intersects the Hugoniot curve, and there are two solutions for a given detonation velocity, namely weak and strong (overdriven) detonations. Similarly, the Rayleigh line intersects with the lower branch giving weak and strong deflagrations. While the upper tangency solution for detonations matches well with experiments, the maximum-velocity solution for deflagrations is typically not observed.

1.7 Detonation structure

Most self-sustaining detonations in gases are inherently unstable and have a three-dimensional non-steady cellular structure [48]. Although these structures lack steady-state characteristics, the experiments have shown that globally they maintain a steady average velocity that is remarkably close to the Chapman–Jouguet (CJ) speed [48]. Detonation instability appears as a pulsation in one-dimensional settings and cellular instability in multi-dimensions [21]. In the following sections, we discuss the one-dimensional structure of detonations using the Zeldovich-von Neumann-Döring (ZND) model and two-dimensional characteristics of detonation front propagation. It is noteworthy that these features have been explored in Publication II and Publication III for the detonation of PRF and PRF-E mixtures with air.
1.7.1 ZND structure of detonations

As discussed in section 1.6, the use of gas-dynamic conservation laws allows the evaluation of the equilibrium states of shock, deflagration, and detonation. However, these conservation laws do not provide information about the transition through the detonation front. Therefore, to describe this transition zone, a model for the structure of the detonation wave must be defined. The model commonly used for the steady 1D planar structure of detonation is the ZND model [48]. Likewise, this model has been applied in Publication II to analyze the 1D planar structure of detonation in PRF and PRF-E mixtures. Figure 1.5 illustrates the ZND detonation structure, depicting normalized pressure ($\frac{P}{P_0}$), temperature ($\frac{T}{T_0}$), and density ($\frac{\rho}{\rho_0}$) obtained for an H$_2$/air mixture at pre-shock conditions of $P_0 = 6670$ pa and $T_0 = 300$ K.

The ZND model describes the 1D detonation front as a shock wave (propagating to the left in Fig. 1.5), leading to adiabatic increases in pressure, temperature, and density behind the shock front (left boundary). Subsequently, the post-shock elevated temperatures and pressures accelerate the induction kinetics producing active radicals within the induction length (time) [48]. These reactions are followed by exothermic reactions which result in rapid HRR and subsequent increase in temperature and drop
in pressure and density. In Fig. 1.5, the exothermic (reaction) zone/time is demonstrated approximately by borders where HRR reaches approximately 5% of its maximum value. Downstream the reaction zone the burnt products further expand (pressure, density drop) until reaching the CJ equilibrium state (right boundary). This expansion generates the forward thrust that sustains the advancement of the leading shock. The ZND detonation structure can be attained by simultaneously integrating a set of 1D steady state Euler equations for mass, momentum, and energy within the reference frame of shock front [48].

The 1D pulsating instability of detonation arises due to the temperature sensitivity of chemical reactions. Even slight fluctuations in shock temperature lead to significant variations in both the induction time and, to a lesser extent, the HRR. Longer reaction times tend to spread out energy release, mitigating the impact of induction time fluctuations. Consequently, longer reaction times possess a stabilizing effect, explicitly accounted for in the stability parameter proposed by Ng [59] as,

\[ \chi = \frac{\Delta_i}{\Delta_e}. \] (1.19)

In the above formula, the stability parameter (\(\chi\)) utilizes the ratio of the induction length (\(\Delta_i\)) to the exothermic length (\(\Delta_e\)), multiplied by the temperature sensitivity in the induction zone (\(\epsilon_i\)), as an indicator of detonation stability [59]. For weakly unstable mixtures, \(\Delta_i/\Delta_e\) is approximately one, yet values as high as 10 have been reported for such detonations [81, 4, 66].

### 1.7.2 Two-dimensional structure of detonations

As previously mentioned, real-world gaseous detonations commonly exhibit a three-dimensional structure sustained by the dynamics of large-scale "cellular" patterns. Such cellular dynamics emerge from the transverse oscillatory nature of the 2D/3D detonation front, which subsequently manifests into longitudinal pulsations along the direction of detonation propagation [48]. The transverse oscillations observed in the 2D detonation front stem from the interaction of two opposing pressure waves, termed transverse waves (TWs), that sweep across the surface of a leading shock front, referred to as the incident shock (IS). Upon collision, these TWs reflect from each other, resulting in the formation of a Mach stem (MS) or reflection. The Mach reflection generates two triple-shock configurations, known as triple points (TPs), positioned on either side of the MS and pushed away by its force. Each triple point comprises a TW, an IS, a MS, and a slip line (SL) located between the TW and the MS. These slip lines further give rise to strong shear layers, separating the MS (or IS) from the TW, as illustrated in Figure 1.6.
The periodic nature of cellular dynamics contributes to the self-sustained propagation of cellular detonations [48]. As the two triple points TPs (TP1) move apart, MS existing between them undergoes a gradual dissipation of its energy. In due course, each of the TP1s collides with an incoming set of TPs (TP2) traveling in the opposite direction, giving rise to a fresh set of TPs (TP3). Consequently, the formerly decaying MS transforms into an IS for these newly approaching TP3s. This intermittent sequence entails the decay of an MS into an IS, which is then succeeded by the emergence of a new MS to replace it, completing the cycle.

Following the collision of TPs, the impulsively generated MS is relatively strong, obtaining a high-pressure gradient and a propagation speed approximately 1.5 times the CJ speed ($U_{CJ}$). As the cycle progresses and the MS decays into an IS, its velocity and pressure gradient gradually diminish, reaching propagation speeds in the order of 0.5 times $U_{CJ}$ before the onset of the subsequent TPs collision [48]. Consequently, within one cycle, the leading shock in the 2D detonation pulsates between the two MS stages of propagation. This dynamic process is visually captured in Figure 1.7, where the case configuration, TP trajectories, and mesh sensitivity analysis for a 2D planar direct-initiated H$_2$/air detonation at $P_0=6670$ pa and $T_0=300$ K are depicted. The cyclic nature becomes apparent within the marked cell II in Figure 1.7-b), evident by the darker-colored first half (indicating a higher pressure gradient) and the lighter-colored second half.
Introduction

of the cell II. It is notable that, each detonation cell effectively corresponds to the footprint of one complete TP collision cycle.

Figure 1.7. 2D planar detonation of H₂/air at P₀=6670 Pa and T₀=300 K, a) schematic presentation of the simulation setup, b) detonation cellular structures inside the channel, c) sensitivity of the triple point trajectories to the grid resolution. Image from Publication III.

Meanwhile, the reactive compressed gas located behind the leading shock undergoes a sequence of events marked by rapid reactions, initiated by the elevated temperature and pressure in the vicinity of the MS and at the TPs. This phase is followed by a comparatively longer IDT during the decaying stage toward an IS, characterized by reduced temperature and pressure [69]. Thus, when observed collectively, the composition of a 2D detonation front involves alternating strong and weak shock waves, each accompanied by distinct phases of rapid and decayed reactions, respectively. Furthermore, within this 2D arrangement, transverse waves and colliding triple points move in a direction perpendicular to the propagation, while slip lines induce strong shear effects behind the primary front.

In cases where mixtures exhibit a high sensitivity of induction kinetics to temperature (εᵢ), the leading shock can dispatch from the reacting zone (exothermic region) due to extended induction times associated with the IS. This can lead to the formation of unreacted gas pockets subsequent to the collision of two TPs. Consequently, a greater number of particles downstream of the front remain active, resulting in a delay in achieving the equilibrium state downstream and a propagation velocity lower than
In later stages, the unreacted gas permeates the slipping and shear layers, undergoing combustion as it progresses. Such scenarios are termed "unstable" detonation front propagation which is also characterized by the resulting "cell irregularity" [69, 23]. For unstable detonations, transverse waves are continuously generated and eliminated, hence giving rise to irregular cell patterns.

In addition to the sensitivity of induction zone kinetics, the influence of curvature and confinement levels are recognized as significant factors affecting detonation stability [60, 22, 69, 21]. The curvature of a shock front induces an additional expansion of the gas behind it, resulting in cooling that can mitigate the chemical reactions [60]. Furthermore, in a broader context, the extent to which a detonation develops its intrinsic characteristics is influenced by the degree of confinement [21].

1.8 Hotspot Ignition Regimes

In Publication II and Publication III, the formation of the ignition front within the end-gas of SI engines is investigated in both 1D and 2D scenarios. This phenomenon typically relies on a favorable reactivity gradient within the mixture, which can lead to intense knocking (pressure oscillation) through the development of detonation. The SWACER mechanism (for “shock wave amplification by coherent energy release”), proposed by Lee et al. [47], explains the rapid transformation of a weak shock from a volumetric explosion into an overdriven detonation at a local hot spot. This mechanism synchronizes the chemical energy release with the traveling compression pulse. To achieve this resonance, an induction time gradient is essential, where the mixture is about to ignite upon the arrival of the compression pulse. The resulting pressure rise reinforces the compression pulse, rapidly amplifying a weak shock into a detonation wave. Notably, Zeldovich et al. [95, 94] also conducted numerical simulations demonstrating a similar coupling between shock and reaction zones in nonuniform, preheated mixtures, potentially relevant to engine knock.

Zeldovich [94] presented a method for determining the propagation speed of an ignition front, denoted as \( S_{ign} \), shown in Eq. 1.5. Auto-ignition gives rise to two distinctive types of waves: a reaction wave representing chemical reactions and a pressure wave (shock wave) generated by the thermal explosion of the mixture (termed reaction shocks). Zeldovich categorized ignition fronts into four distinct groups based on their propagation speeds, including deflagration, subsonic ignition, supersonic ignition, and detonation. The critical temperature gradient \( \nabla T_c \) that pertains to this process is defined as:

\[
\nabla T_c = (aU_a)^{-1},
\]
with $U_a$ representing the local speed of sound. Building upon the Zeldovich theory, Bradley and his colleagues extended the analysis to examine ignition regimes within a spherical hotspot. A diagram, referred to as the "detonation peninsula," has been constructed to map out these regimes based on two dimensionless parameters, $\xi$ and $\epsilon$ \cite{7, 20}, where:

$$\xi = \frac{U_a}{S_{ign}} = \frac{\nabla T}{\nabla T_c}, \quad (1.21)$$

and

$$\epsilon = \frac{r_h/U_a}{\tau_e}. \quad (1.22)$$

Here, $\tau_e$ stands for the excitation time, and $r_h$ represents the hotspot radius. The parameter $\xi$ characterizes the coupling between the acoustic wave and the reaction front, while $\epsilon$ indicates the ratio of the chemical timescale to the acoustic timescale within the hotspot. The detonation peninsula involves the range of $\xi$ values that lead to detonation initiation within a hotspot. Moreover, $\epsilon$ quantifies the rate at which chemical energy is transferred to the pressure wave. Notably, the values of $\tau$, $\tau_e$, $U_a$, and $\alpha$ are established at a reference temperature ($T_{ref}$), and the choice of $T_{ref}$ can influence $\xi$ and $\epsilon$. In this investigation, $T_{ave} = T(r = r_h/2)$ is designated as $T_{ref}$ to ensure consistency with the original peninsula concept introduced by Bradley et al. \cite{7}. The 0D constant volume reactor model is employed to compute $\tau$ values, which, in turn, yield estimates for $\alpha$. In Publication II, $\tau_e$ is defined as the time taken to reach 5 percent of the maximum heat release in the constant volume reactor simulations.

Figure 1.8 illustrates various ignition regimes of PRF/air mixture from Publication III which was reconstructed from our 1D simulations in Publication II. The initial pressure is relatively high $P = 50$ bar and the average temperature is in the NTC range, i.e. $T_{ave} = 900$ K. The diagram showcases zones of sub-supersonic ignition in blue and pale blue, while the red area represents the detonation zone. These zones are outlined based on hotspot pressure levels, segmenting the diagram into two regions of relatively high and low pressures, utilizing a predetermined threshold.
As a result, regions surpassing the threshold are designated as detonation zone, while the remaining areas are categorized into supersonic and subsonic ignition regions. In Fig. 1.8, the bottom-left blue zone corresponds to supersonic ignition, whereas the top-right blue zone signifies subsonic ignition. The graph highlights the distinction between the original peninsula and the estimated peninsula from our simulations due to the sensitivity of the regime diagram to fuel chemistry [76, 13, 74, 65, 17] and the utilized chemical mechanism [85, 49]. Furthermore, the green and purple dash-dotted lines delineate phenomena arising from the NTC behavior of the PRF mixture, termed off-centered ignition and coolspots. Additional information about these phenomena is available in sections 3.2 and 3.3, with more detailed insights provided in Publication II and Publication III.

1.9 Objectives of the dissertation

In this dissertation, various temperature stratification-induced combustion regimes associated with abnormal combustion in SI engines are numerically investigated in order to obtain a better understanding of the thermo-physical and thermo-chemical conditions responsible for knocking.
In Publication I, a diagnostic approach (theory and 1D simulations) is presented to assess the prevalence of deflagration and spontaneous ignition as secondary combustion modes for two gasoline surrogates (PRF and PRF-E) under engine-relevant temperatures and pressures. Building on this, Publication II investigates the same fuels under identical average temperatures and pressures, further expanding 1D investigations to explore various ignition regimes and reconstruct the Bradley detonation peninsula for the gasoline surrogates. Lastly, Publication III introduces an efficient numerical framework and extends previous studies to 2D hotspot ignition scenarios, examining curvature effects on shock/detonation fronts, front propagation beyond the hotspot border, and pressure wave reflections from solid walls.

The main objectives of this dissertation are:

1. Analyze the effect of temperature stratification on the prevalence of deflagration vs. spontaneous ignition of gasoline surrogates (PRF and PRF-E) across a range of engine-relevant pressures and temperatures. Construct regime diagrams based on 1D simulations to evaluate combustion characteristics and formulate a diagnostic approach (including 1D regime diagrams and $\beta$-curve theory) enabling prediction of the two combustion modes (Publication I).

2. Examine 1D hotspot-ignition induced detonation, subsonic, and supersonic ignition for gasoline surrogates (PRF and PRF-E), reconstructing detonation peninsula under various engine relevant pressures and temperatures. Furthermore, explore the influence of NTC chemistry on ignition modes and their propagation patterns inside the hotspot (Publication II).

3. Investigate 2D hotspot ignition within a cylindrical setup, and study ignition front propagation and development extending beyond the hotspot region. Furthermore, compare ignition-front patterns between 1D and 2D simulations to identify differences and similarities (Publication III).

4. Develop the required numerical framework for the simulation of hotspot ignition regimes in OpenFOAM in order to handle both accuracy and computational efficiency (Publication II & Publication III).

1.10 Outline of the dissertation

In this section, we have provided a brief overview of the research question pertaining to knocking in SI engines, engine-relevant metrics and correlations, and the theory and specifications related to distinct combustion regimes that are relevant to engine knocking. In Chapter 2, we will delve into the governing equations and the numerical methodology employed for simulations in Publication I - Publication III. Following this, Chapter 3 offers a summary of the results from Publication I - Publication III. Finally, Chapter 4 proceeds with a brief conclusion, highlights on acknowledged
Introduction

limitations, and a recommendation for future work. In addition, the code snippes associated with the developed code are outlined in Appendix A.
2. Methodology

The case studies in this work involve a range of scenarios with reacting flows characterized by both relatively low speeds with gradual pressure variations and high speeds (Mach > 0.3) featuring rapid pressure oscillations (shock waves). As a consequence, in the present work, we employ both low Mach number and compressible flow assumptions [67]. Notably, compressibility effects are typically associated with high-speed gas flows in which density changes resulting from pressure variations significantly impact the flow throughout the field [78]. For CFD analysis of these reacting flows, the open-source toolkit OpenFOAM has been employed. The subsequent sections delve into a comprehensive discussion of the assumptions and governing equations that are employed in our studies.

2.1 Governing equations

The formulation of the governing equations for the flow field is based on the Navier-Stokes (N-S) equations, which include continuity, momentum, and enthalpy equations. These equations are coupled with species transport equations, incorporating chemical source terms calculated using the finite rate chemistry assumption:

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho u) = 0, \tag{2.1}
\]

\[
\frac{\partial (\rho u)}{\partial t} + \nabla \cdot (\rho uu) = \nabla \cdot \tau - \nabla p, \tag{2.2}
\]

\[
\frac{\partial (\rho Y_k)}{\partial t} + \nabla \cdot (\rho u Y_k) = \nabla \cdot (\rho D \nabla Y_k) + \dot{\omega}_k \quad (k = 1, \ldots, N_{sp} - 1), \tag{2.3}
\]

\[
\frac{\partial (\rho h)}{\partial t} + \nabla \cdot (\rho uh) - \frac{\partial p}{\partial t} = \nabla \cdot (\rho \alpha_t \nabla h_s) + \dot{\omega}_h. \tag{2.4}
\]

In the equations above, the symbols \(\rho, u, p, Y_k, h, h_s, \tau, \alpha_t,\) and \(D\) represent the variables for density, velocity, pressure, mass fraction of species \(k,\) total and sensible enthalpy, viscous stress tensor, mass, and thermal diffusivities, respectively. Here, \(N_{sp}\) denotes the total number
of species present in the mixture composition. It is important to note that in OpenFOAM, to calculate species mass fractions, a total of \( N-1 \) equations are solved, with the mass fraction of the last (typically inert) species calculated using \( 1 - \sum_{1 \leq k \leq N_{sp} - 1} Y_k \). The production rates for each species and the HRR are denoted as \( \dot{\omega}_k \) and \( \dot{\omega}_h \), where \( \dot{\omega}_h = \sum_k \Delta h^0_{f,k} \dot{\omega}_k \), with \( \Delta h^0_{f,k} \) representing the enthalpy of formation.

It is notable that, the present work utilizes the unity Lewis number assumption \( (D = \alpha_t) \). This choice is due to the fact that the estimated effective Lewis number \([26]\) for the PRF and PRF-E/air mixtures investigated in Publication I and Publication II is approximately unity. This suggests that the magnitudes of diffusivity and thermal conductivity are comparable, making the unity Lewis number assumption suitable for their characteristics. Moreover, in Publication III, the unity Lewis assumption is adopted for simulations of directly initiated \( \text{H}_2/\text{air} \) detonations. This choice is based on previous findings that, in the absence of strong instabilities, the influence of molecular diffusivity tends to be negligible \([23, 52, 62, 63]\). Additionally, viscosity and thermal conductivity are calculated using the Sutherland law and the Eucken approximation, respectively \([68]\). The ideal gas law and thermal equation of state are also employed to close the system of equations.

2.1.1 Pressure based solution

In Publication I, we studied the prevalence of spontaneous ignition versus deflagration in the end-gas. In the absence of strong compressibility effects, a low Mach number assumption was utilized (Mach < 0.3). Such an assumption holds reasonably well during deflagration simulations, where pressure perturbations resulting from energy release in the reaction zone (manifesting as acoustic waves) have negligible effects on the flow. Moreover, the flame propagation speed \( (S_f) \) is significantly slower than the speed of sound \( (a) \), i.e., \( S_f \ll a \) \([67, 94]\). This leads to acoustic waves equalizing pressure levels within the domain before flame propagation, resulting in heat release occurring at approximately uniform pressure. However, the situation may vary during spontaneous ignition. Rapid reactions elevate the thermodynamic pressure to that of a constant-volume reactor \( (P_{cv}) \) in the burnt products. Yet, the ignition front propagation speed \( (S_{ign}) \) could be orders of magnitude greater than the speed of sound, i.e., \( S_{ign} \gg a \) \([94]\). Consequently, pressure perturbations stemming from gas expansion in the burnt products do not significantly impact the mixture ahead of the reaction front, hence exerting a minimal effect on the dynamic pressure throughout the simulation. Consequently, for cases involving deflagration and spontaneous ignition, we may employ a pressure correction equation in conjunction with the ideal gas law to update the thermodynamic pressure.

Given the above considerations in Publication I, a pressure-based solver
termed reactingFoam was utilized for the simulations. The solver has been utilized previously in a number of studies involving spontaneous ignition and deflagration [40, 30, 58]. The OpenFOAM pressure-velocity algorithms, as discussed in [1], can be demonstrated using the simplified continuity equation for incompressible, inviscid flow:

$$\nabla \cdot u = 0.$$  \hspace{1cm} (2.5)

Furthermore, the momentum equation can be discretized into a set of algebraic equations:

$$M[u] = -\nabla p.$$  \hspace{1cm} (2.6)

Here, the matrix $M[u]$ can be decomposed into diagonal and off-diagonal elements:

$$M[u] = Au - H.$$  \hspace{1cm} (2.7)

Substituting Eq. 2.6 yields:

$$Au - H = -\nabla p.$$  \hspace{1cm} (2.8)

Rearranging Eq. 2.8 leads to the velocity correction equation:

$$u = \frac{H}{A} - \frac{1}{A} \nabla p.$$  \hspace{1cm} (2.9)

By interpolating $u$ to the faces and multiplying by the face area vector $S_f$, the volumetric flux is given by:

$$\phi = u_f \cdot S_f = \left( \frac{H}{A} \right)_f \cdot S_f - \left( \frac{1}{A} \right)_f S_f \nabla p.$$  \hspace{1cm} (2.10)

Applying discretization of the continuity equation ($\nabla \cdot \phi = 0$), from Eq. 2.5, to 2.10 yields the pressure equation:

$$\nabla \cdot \left[ \left( \frac{1}{A} \right)_f \nabla p \right] = \nabla \cdot \left( \frac{H}{A} \right)_f $$  \hspace{1cm} (2.11)

It is noteworthy that in the context of reactingFoam, a modified version of Eq. 2.11 is employed to take into account larger pressure variations:

$$\frac{\partial p}{\partial t} - \nabla \cdot \left[ \left( \frac{1}{A} \right)_f \nabla p \right] + \nabla \cdot \left( \frac{H}{A} \right)_f = 0$$  \hspace{1cm} (2.12)

### 2.1.2 Density based solution

In line with the primary objectives of this dissertation, as outlined in Publication II and Publication III, where we investigate pressure oscillations generated by hotspot ignition, we developed a density-based solver using rhoCentralFoam [19] to solve the governing flow equations. These equations are elaborated upon in section 2.1. As part of OpenFOAM,
rhoCentralFoam utilizes a second-order central, Kurganov and Tadmor (KT) [44] discretization scheme tailored for capturing shock waves within high-speed non-reacting flows. In Publication II and Publication III, temporal integration in the newly developed solver was adapted to the second-order explicit Runge-Kutta (RK) scheme. Additionally, similar to the approach in Ref. [42], the solver incorporated species transport equations along with chemical reaction source terms. As mentioned earlier, compressible fluid dynamics involves the transport of properties not only by the flow itself but also through the propagation of waves. Such a condition necessitates the development of convective flux interpolations that account for the possibility of convection with speeds resembling the local speed of sound. In the next section, we delve into such considerations, particularly focusing on the methodological details of the KT approach.

Kurganov and Tadmor Method

The convective terms within the conservation equations, as outlined in section 2.1 are expressed as $\nabla \cdot (\rho u), \nabla \cdot (\rho uu), \nabla \cdot (\rho uh), \nabla \cdot (\rho uY)$. In the OpenFOAM framework, each of these terms is integrated over a control volume and linearized as follows:

$$\int_V \nabla \cdot [u \Psi] dV = \sum_f [S_f \cdot u_f] \Psi_f \approx \sum_f \phi_f \Psi_f. \quad (2.13)$$

Where subscript $f$ denotes interpolated variables on the cell faces, and $\Psi$ denote conserved variables i.e. $\rho, \rho u, \rho h, \rho Y$ and $\phi = S_f \cdot u$ represent the volumetric flux. The rightmost term in Eq. 2.13 is calculated by splitting the convective term (flux) into flow outward (e.g. superscript $+$) and inward (e.g. superscript $-$) of the face owner cell, utilizing the KT central scheme as below:

$$\sum_f \phi_f \Psi_f = \sum_f \left[ \alpha \phi_f^{+} \Psi_f^{+} + (1 - \alpha) \phi_f^{-} \Psi_f^{-} + \omega_f (\Psi_f^{-} - \Psi_f^{+}) \right]. \quad (2.14)$$

The first two terms, i.e. $\alpha \phi_f^{+} \Psi_f^{+}$ and $(1 - \alpha) \phi_f^{-} \Psi_f^{-}$, correspond to fluxes interpolated in the $f+$ and $f-$ directions on cell faces, respectively. These terms are equally weighted with a coefficient $\alpha=0.5$, reflecting the central nature of the KT scheme [44]. Furthermore, the interpolated conserved variables ($\Psi_f^{\pm}$) in Eq. 2.14 are calculated by using total variation diminishing (TVD) limiters (e.g. Minmod and van Leer, van Albada [93, 28]) for instance:

$$\Psi_f^{+} = g_{f+} \Psi_N + (1 - g_{f+}) \Psi_P, \quad (2.15)$$

where coefficient $g_{f+} = \beta (1 - k_f)$, $k_f$ is a weighting coefficient, and $\beta$ is defined separately for each TVD limiter (details are mentioned in [19]). Moreover, the subscripts $P$ and $N$ indicate the face owner cell and the
neighboring cell, respectively. The third term in Eq. 2.14 introduces an additional diffusion term when the convective term is part of the material derivative (e.g. \( \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho u) \)). The diffusive term involves both maximum volumetric fluxes \( \omega \) and TVD limiters \( (\Psi_f - \Psi_f^+) \) contribution to stabilize the central scheme solution. The value of \( \omega \) is calculated as follows:

\[
\omega = \frac{1}{2} \max(\varphi_f^+, \varphi_f^-). \tag{2.16}
\]

The volumetric fluxes including the local speed of waves (note the difference between \( \varphi \) and \( \phi \)) are calculated in the \( f^+ \) and \( f^- \) directions:

\[
\varphi_f^+ = \max \left( c_f^+ |S_f| + \phi_f^+ c_f^+ |S_f| + \phi_f^-, 0 \right), \tag{2.17}
\]

\[
\varphi_f^- = \max \left( c_f^- |S_f| - \phi_f^+ c_f^- |S_f| - \phi_f^-, 0 \right). \tag{2.18}
\]

Here \( c_{f \pm} = \sqrt{\gamma RT_{f \pm}} \) indicate the local speed of sound (waves), which outlines the inclusion of transport through the wave propagation mechanism in the KT scheme.

### 2.2 Finite rate chemistry solution

Reactions can exhibit both spatial and temporal evolution, i.e. reaction fronts (e.g. deflagration or detonation), or occur entirely uniformly with only temporal changes, e.g. homogeneous reactors. Nonetheless, the reaction zone undergoes two primary stages. The initial stage involves induction kinetics, responsible for generating active radicals, that are thermally neutral or endothermic [88, 48, 24]. The subsequent stage encompasses rapid exothermic reactions where the chemical energy release takes place. This second stage transpires rapidly within the high-temperature regions of the reaction front and typically correlates linearly with temperature [48]. On the contrary, the first stage, often referred to as the ignition delay or induction stage, is relatively slower (at typical operating temperatures) and exhibits a logarithmic and non-linear (e.g. hydrocarbons with low/intermediate-temperature chemistry) correlation with temperature. Hence, in combustion scenarios where the first stage holds significance, the assumption of finite rate chemistry becomes paramount.

In CFD of reacting flows, the finite rate chemistry assumption often necessitates the inclusion of dozens of species and hundreds of reactions within the chemical kinetic mechanism. Crucially, it necessitates solving the system of kinetic ODEs, at least once while solving the governing equations of the flow field. Given the substantial computational load imposed by this ODE solution, simplified (skeletal) chemical kinetics, involving a reduced number of species, is commonly employed in CFD simulations [61, 73]. Nevertheless, the solution of chemical kinetics ODEs
remains a bottleneck in the majority of reacting CFD scenarios. It is worth noting that the selection of the chemical model used for deriving chemical source terms is tied to the nature of the combustion (constant or variable pressure) process and the coupling between the Navier-Stokes and ODE solutions. In the following sections, we explain the utilized chemical model and introduce the numerical method employed for ODE solution.

2.2.1 Chemical source term evaluation

For simulating reacting flows, OpenFOAM employs a first-order operator splitting scheme [51, 43] to evolve the flow field and thermochemical composition due to reactions over a time step, $\Delta t$. During the solution of the chemical source terms in Eqs. 2.4 and 2.3 ($\dot{\omega}_k$, $\dot{\omega}_h$), the flow field characteristics are assumed to be approximately constant in both space and time [64]. As discussed in section 2.1.1, since flame speeds are significantly smaller than the speed of sound, heat release is considered to occur at a constant pressure [67, 94]. Consequently, the governing equations solved during the reaction sub-step correspond to a constant-pressure (CP) homogeneous reactor:

$$\frac{\partial T}{\partial t} = -\frac{1}{\rho c_p} \sum_{k=1}^{N_{sp}} h_k W_k \dot{\omega}_k,$$

(2.19)

$$\frac{\partial Y_k}{\partial t} = \frac{W_k}{\rho} \frac{\dot{\omega}_k}{\rho} (k = 1, ..., N_{sp} - 1),$$

(2.20)

Here, $c_p$ represents the constant-pressure mixture-averaged heat coefficient, $h_k$ denotes the enthalpy of the kth species, and $\dot{\omega}_k$ stands for the production rate. In CFD simulation of reacting flows, the constant-pressure assumption implies that the pressure remains unchanged during the reaction substep of an operator splitting scheme. It is important to clarify that this assumption does not imply a fixed pressure throughout the entire simulation but is particularly applicable to the reaction stage. This assumption is frequently used in simulations of low-speed combustion scenarios [67].

The choice of a first-order operator-splitting method arises from the resource-intensive nature of solving Eqs. 2.19 and 2.20 when employing detailed or skeletal chemical kinetics mechanisms [73, 64]. Notably, a 115-species skeletal toluene primary reference fuels (TPRF) mechanism by Stagni et al. [84] was employed for PRF simulations in Publication I, Publication II, and Publication III. For PRF-E mixture simulations, a 171-species skeletal bio-gasoline mechanism [71] was utilized, in Publication I and Publication II, and for the directly initiated H$_2$/air detonation simulations in Publication III, the Marinov et al. [54] mechanism with 10 species and 27 reactions was chosen.
2.2.2 Integration of kinetic ODEs

Detailed and skeletal chemical mechanisms may include a wide range of reactions often with varying timescales (that can be as small as $10^{-12}$ s), making their modeling computationally stiff [50]. To address this challenge, reactive-flow simulations often utilize implicit algorithms for integrating the stiff equations governing chemical kinetics, as seen in Eqs. 2.19 and 2.20. Implicit methods require frequent evaluations of the Jacobian matrix [50, 18], which can be numerically approximated using finite difference methods. However, for larger chemical kinetic models, this approach can impose a significant computational load since the number of evaluations scales with the square of the species count. Nevertheless, by utilizing an analytical formulation for the Jacobian matrix rather than relying on the conventional finite difference approximation, the computational expense associated with numerous evaluations can be reduced from a quadratic scaling to a linear one [61].

In Publication I-Publication III, the chemical source terms are calculated using direct integration of finite rate chemistry. The Jacobian matrix for the system of kinetic ODEs, Eqs. 2.19 and 2.20, is calculated analytically using pyJac [61] generated routines. These routines are further incorporated in CFD simulations and using its recent implementation to the OpenFOAM framework, DLBFoam v.1.1 [58]. For further details on the implementation and improvements over the standard OpenFOAM solvers, the reader is referred to [30, 58, 87]. It is important to highlight that the ODE formulation of kinetic equations renders these operations within a single cell isolated from neighboring cells during the reaction substep in CFD simulations. This distinctive characteristic will be shown to facilitate the load balancing of chemical computations during CFD simulation.

Notably, the constant volume (CV) variation of the aforementioned ODE system solution has been tested for 1D direct initiation of detonation in an H$_2$/air mixture. The comparative analysis demonstrates that the reaction rates calculated using CV and CP reactors exhibit marginal differences, owing to the utilization of very short time steps. These minor discrepancies are not expected to significantly alter the overall combustion dynamics, particularly in scenarios involving varying pressures. Furthermore, the validation of the CP reactor assumption in the solver is showcased through simulations of CH$_4$ and hydrogen-air mixture detonation in Publication II and Ref. [80]. These validations involve comparisons with simulations conducted by Marcantoni et al. [53] and Pan et al. [65].
2.3 Simulation efficiency improvement

In the OpenFOAM framework, similar to most CFD codes, parallelization is achieved through geometrical domain decomposition [28, 93] prior to simulation start. Nevertheless, several challenges can manifest as bottlenecks within parallel reacting flow simulations, primarily stemming from the localized distribution of reaction zones (in flame/ignition fronts) within the computational domain. This distribution, coupled with the intensive computational demands of these reacting regions, often results in an unequal distribution of computational load among different processors in the pre-discretized domain. Consequently, addressing this issue holds significant potential to enhance simulation efficiency.

In Publication III, we focused on studying the 2D hotspot ignition of PRF/air mixture, specifically exploring sub-supersonic ignition fronts and detonation propagation under conditions relevant to SI engines. To accomplish this, we developed a comprehensive numerical framework that incorporates efficient simulation techniques, including adaptive mesh refinement (AMR), mesh balancing, and chemistry problem balancing. These techniques were integrated into our density-based solver (section 2.1.2) to enhance its performance. The resulting numerical framework, termed ARCFoam (ARC=Adaptive, Runge Kutta, Central), was utilized for various case setups, such as the direct initiation of H₂/air detonation and PRF hotspot ignition.

In the subsequent sections, we delve into the different numerical techniques employed in Publication III to improve the efficiency of our simulations. Figure 2.1 presents an overview of the flowchart outlining the sequence of processes within each timestep during run-time in ARCFoam. The flowchart illustrates key stages, including mesh pre-processing labeled as "Mesh Update and Balance," as well as the sequence of density-based solver procedures (flux calculation and flow variable updating) executed through Runge-Kutta (RK) iteration. The RK iterations also include updating chemistry through the utilization of DLBFoam v.1.1. [87, 58].
Methodology

2.3.1 Adaptive Mesh Refinement

OpenFOAM employs an h-adaptivity technique known as Adaptive Mesh Refinement (AMR) [28] to enhance simulation efficiency. This strategy selectively enhances mesh resolution in relevant regions, like flame and shock fronts, while maintaining lower resolution in adjacent parts of the flow [14, 96]. The AMR effectiveness is justified by the reduced number of cell counts compared to the fully refined computational domain during the simulations. Initially designed for 3D hexahedral meshes, the AMR capability of OpenFOAM (referred to as OF-AMR hereafter) has been extended for 2D scenarios in recent years by the community [41, 6, 82]. In Publication III, we utilized the 2D OF-AMR for simulations involving $\text{H}_2$
and PRF mixtures with air. Notably, this method was further enhanced to incorporate mesh balancing, a modification aimed at improving simulation efficiency (refer to section 2.3.2). Figure 2.2-a illustrates a schematic of a two-level refined grid in 2D scenarios, highlighting two refinement regions marked by blue/purple ($w_1/w_2$) and yellow/red ($w'_1/w'_2$) colors. The variables $w$ and $w'$ serve as indicators of the local weight (reflecting the number of cells) within each cluster, a concept elucidated in section 2.3.2.

![Figure 2.2](image)

**Figure 2.2.** Schematic demonstration of a) visible cells in the two refinement regions, ($w_1,w_2$) and ($w'_1,w'_2$) indicate local weights (number of cells) of each refinement region, b) different cell levels (1 and 2) on each refinement region and their projection on parent cells (level 0), c) mesh and temperature distribution of Richtmyer–Meshkov instability in H$_2$ detonation with 5 levels of refinement, image from *Publication III*.

In the first step of OF-AMR, a refinement criterion (normalized error function) is computed across the domain. For instance, in *Publication III* we utilized the ratio of second-order to first-order derivative terms in the Taylor expansion of density [97, 86] as the refinement criterion. Next, cells are marked for refinement or unrefinement based on user-defined thresholds (`refineLevel` and `unrefineLevel`). In the subsequent step of 2D refinement, parent cells (with level = 0), as shown in Fig. 2.2 b, are subdivided into four smaller child cells, except in directions normal to empty boundaries. New child cells are assigned a level higher (+1) than their parents. Finally, marked refined cells are then merged to re-integrate into coarser cells during the unrefinement step. The unrefinement process in OF-AMR uses a "tree" data structure (`refinementHistory`) to record refinement information (tree) and manage cell removal. An example of 2D AMR, involving five refinement levels, is shown in Fig. 2.2-c), illustrating mesh and temperature distribution for the Richtmyer–Meshkov instability [98] in the H$_2$ detonation simulation from *Publication III*.

### 2.3.2 AMR mesh balancing

OpenFOAM version 10 [92] introduced Dynamic Load Balancing (DLB) as a strategy to optimize computational efficiency in parallel simulations, featuring a new framework for dynamic load balancing, decomposition, and redistribution. This framework, hereafter referred to as OF-DLB, allows users to calculate the CPU load on each processor during parallel runs.
The loadBalancer translates CPU loads into cell-based weights, utilized by the distributor, i.e. zoltan [15] or scotch [10], to dynamically redistribute the mesh based on cell CPU loads, enhancing solver performance.

In AMR, local cell clusters are commonly generated within the domain, as seen in Figure 2.2. These clusters can significantly increase the computation time of the processors responsible for that particular part of the domain. This situation can greatly reduce the efficiency of the solver, especially when dealing with a large number of cells. To address this, in Publication III, the authors presented a new loadbalancer tailored to the OF-AMR and OF-DLB integrated into ARCFoam. The new loadbalancer enables the distribution of refinementHistory within parallel simulation with OF-AMR. Following the Voskuilen [89] solution, the new loadbalancer ensures parent cells are fully present on each processor, preserving refinementHistory integrity during parallel execution.

The AMR mesh-balancing process includes the following steps. Initially, "visible cells" including the child cells and unrefined parent cells are counted for each processor, and stored as weights. This process is visually demonstrated in Figure 2.2-a), showcasing cell clusters, visible cells, and their corresponding weights (i.e. $w_s$ and $w'_s$). These weights are subsequently projected onto coarse cell maps, illustrated in Figure 2.2-b). In the next step, coarse cell maps alongside their associated weights are sent to the distributor (such as zoltan or scotch) to generate a new balanced cell distribution for the processors. Finally, the cells and their corresponding flow field information are exchanged among processors. This step not only manages the creation of new boundaries for updated cell maps but also ensures the distribution of the refinement tree to host processors. The formulated distribution of the refinementHistory, thereby ensures the integrity of the refinement information on all processors.

2.3.3 Chemistry problem balancing

In scenarios involving reacting flows like flames and detonation fronts, rapid reactions take place within narrow regions. These reactions occur in a small fraction of cells compared to the surrounding cells in the domain, leading to an uneven computational load between processors due to the resource-intensive nature of the chemistry solution. The higher the number of species and reactions in the chemical mechanism (stiffer chemistry solution) the higher the difference between the computational times in these two parts of the domain. However, as indicated in section 2.2, solving chemistry occurs independently on a per-cell basis in homogeneous reaction systems in OpenFOAM. The open-source chemistry update model named DLBFoam [87] takes advantage of this feature and dynamically distributes kinetic ODE problems to available processors during the chemistry update stage, effectively balancing the chemistry workload across resources.
The recent developments in the DLBFoam library [87, 58] can be accessed through the open-source code available at https://github.com/Aalto-CFD/DLBFoam. Earlier code implementations were carried out by Kahila et al. [30], with further details provided in [29]. In our work (Publication I-Publication III), we employed DLBFoam v.1.1, which integrates analytical Jacobian routines generated by pyJac [61] to solve the kinetic ODE system. The core steps of DLBFoam are outlined in the yellow box of Fig. 2.1.

The process in DLBFoam begins by balancing the chemistry problems based on the chemistry solution time of each cell from the previous time step. Each processor handles a cluster of these chemistry problems. After calculating the cumulative chemistry time on each processor, the average is determined, and processors exceeding this average send their surplus chemistry problems to those below the average. Subsequently, the newly balanced chemistry problems are solved on each processor, and cumulative and individual solution times are recorded, completing the cycle. In the final step, these chemistry solutions are returned to their original cell locations, and the flow solution resumes.
3. Summary of publications

3.1 Publication I

The main focus in Publication I is to investigate the relative importance of spontaneous ignition versus deflagration of gasoline surrogates in a thermally stratified mixture under conditions relevant to SI engines. Initially, two gasoline surrogates, namely PRF (RON/MON = 87) and PRF-E (RON/MON = 89/95), were selected. To account for different engine operating conditions, two pressure levels (P) of 20 and 50 bar were chosen, representing normal and boosted engine conditions, respectively. Additionally, three average temperatures ($T_{ave}$) of 830, 900, and 1000 K were considered to encompass relatively low, intermediate, and high end-gas temperatures in SI engines.

To conduct the theoretical (β-curve theory detailed in section 1.5.2) and numerical studies, a one-dimensional domain with uniform pressure and mixture composition (PRF or PRF-E) and initial sinusoidal temperature distribution was employed, with periodic boundary conditions in both theoretical and numerical analyses. The variation of the temperature non-uniformity and length/time scales were selected according to the previous theoretical [40] and large eddy simulation results [55] by the authors. For 1D simulations, a pressure-based solution of the N-S equations coupled with species transport equations and a PIMPLE algorithm for pressure-velocity coupling were utilized. The chemical source terms were calculated using the constant pressure reactor assumption with the DLBFoam v.1.1 [58] modified chemistry solver, see section 2.3.3. In order to account for the impact of low, intermediate, and high-temperature chemistry in the theoretical and numerical analyses, finite-rate chemistry was incorporated. Specifically, a 115-species skeletal TPRF mechanism developed by Stagni et al. [84] was used for PRF simulations, while a skeletal 171-species bio-gasoline mechanism [71] was selected for the PRF-E simulations.

The investigation focused on temperature stratification, varying the si-
nusoidal temperature amplitude ($\delta$) and wavelength ($\lambda$) at selected $T_{ave}$ and P combinations, see section 1.5.2 for details. Figure 3.1 illustrates temperature initialization and subsequent processes. The sinusoidal temperature distribution creates two distinct regions of low and high reactivity (step I, Fig. 3.1), assuming a monotonous correlation between temperature and IDT. Prior to auto-ignition, thermal diffusion reduces temperature differences within the domain. After the induction time, rapid temperature rise due to exothermic reactions occurs (step II, Fig. 3.1). Subsequently, the reactive domain may experience two scenarios: under lower reactivity stratification, the lower portion undergoes secondary ignition after the primary ignition, rapidly burning the rest of the mixture (step III-a, Fig. 3.1). Conversely, under relatively high reactivity stratification, a flame front forms from the primary ignition, burning the remaining mixture similar to a premixed flame (step III-b, Fig. 3.1). Notably, the initial heat diffusion from the primary ignition may impact the temporal reactivity evolution in the lower reactive side.

Figure 3.1. Schematic representation of the combustion process: step I - Thermal diffusion, step II - Primary ignition, step III-a - Spontaneous ignition mode with an ignition front propagating towards the unburnt mixture, and step III-b - Deflagration mode with a premixed flame propagating towards the unburnt mixture.

Furthermore, the low and intermediate heat release of the fuels, which result in NTC behavior, introduces an extra variable dependent on $T_{ave}$ and P, further influencing the aforementioned steps. Figure 3.2 shows how cases I-V are located on IDT versus temperature graphs at 20 and 50 bar, considering both PRF and PRF-E mixtures. As shown in Fig. 3.2-a and b, cases II, IV, and V are located in regions with NTC behavior. Particularly, the NTC behavior can potentially impact the dynamics of ignition front
propagation in the mentioned scenarios by influencing the initial reactivity distribution and interacting with processes like thermal diffusion.

![Figure 3.2. PRF and PRF-E ignition delay time versus temperature profiles at, a: P = 20 and b: 50 bar; black circles are marking the locations of the studied cases.](image)

The key findings from this study are as follows:

1. The combustion regime diagrams (e.g. Figure 3.3) constructed from 1D simulations demonstrate that the presence of fuels’ negative temperature coefficient (NTC) significantly increases the dominance of the spontaneous ignition regime under different levels of temperature stratification at various $T_{ave}$ and P levels.
2. The dominance of spontaneous ignition through NTC manifests at two distinct levels. First, NTC decreases the local reactivity stratification ($\alpha$), leading to an increased spontaneous ignition speed ($S_{\text{ign}}$). Second, NTC contributes to a decrease in global reactivity stratification, resulting in regions with similar IDTs. This phenomenon raises the chances of individual ignitions occurring within the mixture.

3. The dominance of the spontaneous ignition regime in the presence of NTC was also evident in the $\beta$-curve analysis. It was observed that the predictability of the $\beta$-curve decreases in the presence of NTC, mainly due to the challenge of finding a clear balance between deflagration and spontaneous ignition ($S_I = S_{\text{ign}}$) when $\alpha$ approaches zero, leading to $\beta$ approaching zero as well, as seen in Eq. 1.10. Moreover, the reduction in global reactivity stratification blurs the clear boundary between modes, leading to a blended mode between deflagration and spontaneous ignition, termed spontaneous ignition-assisted deflagration in Publication I.

4. Finally, the ONs (RON and MON) of the mixtures correlated with their NTC levels at different pressures through the generated regime diagrams. The PRF-E mixture, with lower NTC behavior, exhibited reduced dominance of the spontaneous ignition mode at higher pressures (RON conditions). However, at lower pressures (MON conditions), both fuels
exhibited notable NTC behavior, Fig. 3.3-c, resulting in a higher prevalence of spontaneous ignition. It was observed that the advantage of lower ignitability associated with the PRF-E mixture was less pronounced under these lower pressure conditions, highlighting the intricate relationship between pressure levels and fuel NTC characteristics, which influence the overall combustion performance and knock resistance of the studied gasoline surrogates.

In summary, these findings offer novel insights into the interactions between 1) thermal diffusion, 2) NTC effects, and 3) temperature stratification, as well as their impact on the prevalence of auto-ignition versus deflagration in the end-gas.

### 3.2 Publication II

Continuing from the findings of the first publication, Publication II provides insights into hotspot ignition regimes (supersonic, subsonic, and detonation) and addresses the knock limits in SI engines, which can lead to different levels of pressure oscillations inside the engine cylinder. This study delves deeper into understanding such distinct ignition behaviors and their potential impact on engine performance and knock severity. To reproduce such scenarios, Bradley's 1D theoretical, see section 1.8, and numerical framework were employed. Building upon the results of Publication I, the same gasoline surrogates (PRF and PRF-E), $T_{\text{ave}}$ (830, 900, 1000 K), and pressure levels (20 and 50 bar) were selected for the theoretical and numerical study. The main objective was to construct 1D hotspot ignition regime diagrams under the mentioned conditions and for the selected gasoline surrogates, emphasizing the influence of average temperature (including NTC range), initial pressure, and ethanol addition on different ignition regimes.

In order to perform 1D hotspot ignition CFD simulations with rapid pressure oscillations, we developed a fully compressible solver that incorporates an explicit formulation of the Navier-Stokes equations, coupled with species transport and a finite-rate chemistry solution (DLBFoam). For detailed information, see section 2.1.2. For the PRF and PRF-E mixture simulations, we selected the 115-species and 856 reactions skeletal TPRF [84] and 171-species and 3751 reactions skeletal bio-gasoline [71] mechanisms which have been validated in Publication I, respectively. To ensure accuracy, we validated the solver through 1D non-reacting shock tube simulations [83] and CH4/air hotspot detonation simulation [65]. Additionally, a mesh sensitivity study on PRF/air mixture hotspot detonation revealed that a 0.75 $\mu$m mesh is sufficient for capturing the ZND peak pressure and achieving mesh convergence. Notably in Publication II, the ZND detonation structures were calculated, using the SDToolbox [39], for the
selected fuels. Moreover, the impact of NTC on hotspot ignition initiation and the propagation patterns of ignition fronts inside the hotspot were explored. Figure 3.4 illustrates different initial temperature profiles and the corresponding propagation patterns inside the hotspot.

Figure 3.4. A schematic presentation of the combustion process shows, (a) hotspot/coolspot initial T profiles, (b) centered ignition and regular propagation inside the hotspot/coolspot, (c) off-centered primary ignition due to NTC and double ignition front propagation, and (d) occurrence of primary ignition and subsequently, formation of the secondary ignition kernel due to NTC

NTC behavior, in particular, can lead to an inverse distribution of ignition delay time (IDT) versus temperature, resulting in higher reactivity at lower temperatures. This phenomenon gives rise to ignition initiation from lower temperatures, which are termed coolspots. Furthermore, NTC can alter the conventional propagation pattern (Fig. 3.4-(b)) by shifting the region of highest reactivity away from the center of the hot/coolspot ($T_{max}/T_{min}$), leading to front propagation in two different directions, i.e. off centered ignition (Fig. 3.4-(c)). Additionally, if the temperature variation within the hotspot spans the NTC zone, there may be multiple individual points with similar IDTs, leading to the appearance of secondary ignition kernels, Fig. 3.4-(d), within the hotspot. These characteristics were noted to significantly impact the dynamics of ignition front propagation within the hotspot and, consequently, the distribution of different ignition modes within the detonation peninsula, as well as the pressure levels within the hotspot.

The key findings from the present numerical study can be summarized as
follows:

1. In general, when comparing pressure oscillations between hotspot simulations and experimental data for mild/normal knock ($\Delta P \approx 5$-24 bar) and superknock ($\Delta P \gtrapprox 50$ bar), it was observed that at elevated pressure conditions (50 bar), superknock might primarily manifest as detonation events. In contrast, normal knocking phenomena appear to be associated with lower pressure levels resulting from spontaneous/supersonic ignition.

![Figure 3.5](image)

**Figure 3.5.** Ignition regime diagram of case I ($P = 50$ bar and $T_{ave} = 900$ K), red zone indicates the detonation peninsula of (a) PRF and (b) PRF-E mixtures, respectively. Colored circles display the hotspot pressure calculated for each 1D simulation. Letters P, B, and N indicate supersonic ignition, subsonic ignition, and the nominal condition for knock initiation inside SI engines [38]. (c) IDT distribution at the initial time versus $T$ of PRF and PRF-E mixtures. The black dashed line represents the original Bradley detonation peninsula.

2. NTC increases the knock propensity by favoring all ignition regimes of the mixture, as depicted in Fig. 3.5 a. However, it also promotes spontaneous/supersonic ignition propagation, which may suppress detonation (superknock) occurrence by limiting the traveling distance required for a developing detonation front.

3. Compared to PRF, PRF-E exhibits lower NTC behavior, as evident in Fig. 3.5c, which in turn leads to a lower knock propensity. This difference can be observed when comparing the detonation zone in Figs. 3.5 a and b. The lower knock propensity of PRF-E can also be attributed to its lower
energy density and longer excitation times. These factors help reduce pressure perturbations and the formation of intense shock waves. Nonetheless, there may be a higher likelihood of experiencing heavy knocking and superknock at the onset of detonation within PRF-E hotspots. The longer excitation times and yet higher tendency toward blended subsonic ignition modes in PRF-E may facilitate the detonation run-up distance and thereby, approaching DDT.

4. Induction time (IDT) was found to have the most significant impact on hotspot pressure levels for the studied gasoline surrogates, compared to energy density and excitation time. The crucial role of IDT can be explained by its direct influence on the dynamics of ignition front propagation, the size of the ignition kernel, and the distance covered by the ignition front during its development.

The novel findings of Publication II can be summarized as follows: 1) Reconstructing the detonation peninsula for gasoline surrogates (PRF and PRF-E). 2) Elaborating the effect of NTC on the prevalence of different ignition regimes on the hotspot ignition regime diagram. 3) Studying non-conventional ignition propagation patterns resulting from NTC within the hotspot.

3.3 Publication III

Building upon the objectives of the two previous papers, Publication III takes the investigation of the knocking phenomenon further into two-dimensional scenarios and explores phenomena beyond the hotspot border. This extension enables a more realistic representation of knocking relevant events, thereby facilitating the examination of 2D phenomena. These include instabilities and interactions between reaction fronts and reflected shock waves. Additionally, the expanded domain of interest enables the study of transitions of the main ignition front within the hotspot boundaries. By comparing the results obtained from the two-dimensional framework with those from the one-dimensional simulations, the study aims to identify both similarities and disparities between the combustion regimes observed in two and one-dimensional settings.

One of the primary challenges in conducting two-dimensional simulations of knocking-related events is the computational expense. In particular, as our previous studies have shown, finite rate chemistry, such as NTC chemistry of gasoline surrogates, plays a significant role in events leading to abnormal combustion. However, applying such a skeletal chemical mechanism in two-dimensional configurations is typically infeasible without special treatments. To address this, Publication III introduces the numerical framework **ARCFoam** (ARC = Adaptive, Runge-Kutta, Central). **ARCFoam** combines dynamic mesh balancing, chemistry balancing (DLBFoam),
and adaptive mesh refinement (AMR), as shown in Fig. 2.2, with the compressible solver introduced and verified in Publication II for high-speed flows.

Publication III explores three different 2D numerical configurations: 2D planar and cylindrical directly initiated detonation of hydrogen/air mixture, as shown in Fig. 3.6, operating under relatively low ambient pressure (0.0667 bar) and temperature (300 K). Additionally, 2D hotspot ignition of the PRF mixture is investigated using selected hotspot conditions from Publication II at $T_{\text{ave}} = 900$ K and $P = 50$ bar. The former simulations are conducted to establish the validity and performance of the solver. Subsequently, we proceed to the more computationally expensive simulations involving the PRF mixture. For the hydrogen/mixture simulations, we chose the chemical mechanism by Marinov et al. [54] with 10 species and 27 reactions. On the other hand, the PRF/air mixture simulations employ the more comprehensive 115-species chemical mechanism (utilized and validated in Publication I), including low/intermediate temperature kinetics, requiring heavier computational resources. To validate and compare the results, corresponding 1D planar simulations were also conducted. In Publication III, we conducted simulations of H$_2$/air mixtures to demonstrate the capabilities and verification of the ARCFoam framework in 2D simulations. These simulations utilized a relatively less computationally expensive 10-species mechanism for modeling the hydrogen/air mixture. In Publication III, the accuracy of the 2D direct initiation of H$_2$/air detonation results has been confirmed through validation against prior numerical investigations, including studies conducted using OpenFOAM with both static mesh [52] and AMR [5] configurations. On the other hand, hotspot ignition of the PRF/air mixture serves as the primary focus, requiring more substantial computational resources due to the utilization of a 115-species chemical mechanism that includes low/intermediate temperature kinetics.
The key results obtained in Publication III can be outlined as follows:

1. The analysis of ignition front characteristics using the 1D Bradley framework showed that, in general, the framework accurately captures the dynamics of the ignition front inside the hotspot in both 1D and 2D simulations. However, certain phenomena, such as the formation of converging/diverging fronts due to the NTC behavior of the fuel or the influence of reflected shock waves generated by the subsonic ignition front, can introduce significant deviations from the predictions of the 1D framework.

2. In the hotspot ignition simulations involving detonation, the process initiates with a rapid burst of spontaneous ignition, followed by a development stage with $S_{\text{ign}}$ lower than the CJ speed, and ultimate transitions to sustained detonation development in both 1D planar and 2D cylindrical settings. However, distinct factors such as variation in initiation energy, cellular instability, and curvature effects lead to significant differences in the initiation and dynamics of the detonation front propagation between the 1D and 2D hotspots with the same initial temperature profiles.

3. A notable effect observed in the 2D cylindrical simulations was the occurrence of converging shock/detonation waves, Fig. 3.7, resulting from off-centered ignition, caused by the NTC behavior of the fuel. These generated converging shock (detonation) waves lead to a drastic increase in temperature and pressure within the hotspot due to the continuous reduction of the shock area. This behavior contrasts with the conventional expectation of diverging shock (detonation) scenarios associated with the typical ignition front propagation in the hotspot.
4. In 2D cylindrical settings, the subsonic ignition front propagation plays a crucial role in the knocking phenomenon. 2D cylindrical simulations depicted that the reflection of shock waves, emanating from the rapid ignition, off the walls resulted in a significant increase in pressure and temperature, followed by secondary ignition. These events, known as shock wave reflection-induced detonation (SWRID) [90], are the most frequently occurring detonation initiations in SI engines and are considered important for understanding realistic engine scenarios.

The novel findings of Publication III can be summarized as follows: 1) A new framework, ARCFoam, designed for efficient simulations of high-speed reacting flows is introduced. 2) 1D framework predictions are valid for the initial ignition mode within the hotspot while NTC and geometrical effects may result in follow-up changes. 3) Unique 2D phenomena are observed in 2D simulations which may not be observed in 1D simulations, including the converging detonation/shock fronts, shockwave reflection induced detonation, and the impact of detonation cellular instability on the transient front structure.
4. Conclusions and perspectives

4.1 Conclusion

In the context of abnormal combustion in SI engines, specifically knocking, numerical investigations were conducted to examine the secondary combustion characteristics of gasoline surrogates under engine-relevant thermal stratification, both in 1D and 2D settings. Despite the limited scope of the current numerical and theoretical study, the secondary (abnormal) combustion occurring within the end-gas region of SI engines displayed a complex topological structure. This included a diverse range of combustion modes, such as spontaneous, subsonic, supersonic ignition, detonation, and deflagration, all influenced by the NTC behavior of the studied gasoline surrogates. Furthermore, the study delved into the potential excessive pressure levels associated with these combustion modes. Additionally, a novel numerical solver and computationally efficient techniques were gathered in a framework, termed ARCFoam based on OpenFOAM, and validated for high-speed reacting flows in both 1D and 2D configurations, as documented in Publication II and Publication III. The following conclusions are drawn from this research:

1. In Publication I, we demonstrated the significant NTC chemistry effect on the balance between spontaneous ignition and deflagration by utilizing the presented diagnostic approach (including 1D regime diagrams and β-curve theory), which shifted the phase boundaries to higher λ and δ values. Furthermore, the diagnostic approach employed in Publication I, exhibited a strong correlation with the octane numbers (RON and MON) of the studied gasoline surrogates (PRF and PRF-E) under relevant temperature and pressure conditions (Objective 1).

2. Focusing on auto-ignition modes in Publication II, we reconstructed 1D hotspot ignition regime diagrams (detonation peninsula) for the gasoline surrogates under identical conditions (T_{ave}, P) as in Publication I. These results highlighted the NTC effect, leading to off-centered ignition,
Conclusions and perspectives

multiple ignition points, and cool spot ignition within the 1D framework. Additionally, it was observed that the increased prevalence of spontaneous/supersonic ignition driven by NTC can prevent detonation development through reduced traveling distance (objective 2).

3. By extending the hotspot ignition simulations to 2D settings in Publication III distinctive 2D physical phenomena were observed, setting the 1D and 2D results apart. These included converging shock/detonation fronts and detonation induced by shock wave reflections. Notably, it was found that while the 1D framework can accurately predict the initial ignition regime within the hotspot, factors such as curvature, geometrical confinement, and NTC effects can lead to significant disparities between the initial prediction and the subsequent characteristics and development of the ignition front (objective 3).

4. During the research documented in Publication II and Publication III, a density-based solver was developed based on rhoCentralFoam. This solver included species transport equations with species and energy reaction source terms. Moreover, the solver was validated against both 1D and 2D reacting problems, as outlined in Publication II and Publication III. Furthermore, in Publication III, this solver was combined with mesh pre-processing methods i.e. OF-AMR and a new mesh balancing method, leading to the creation of a comprehensive numerical framework named ARCFoam. The performance of this integrated framework was also demonstrated in Publication III in 2D direct initiation of H\textsubscript{2}/air detonation simulations (objective 4).

4.2 Limitations

The present dissertation provides valuable insights on the interaction between NTC behavior of gasoline surrogates, pressure wave generation, the dynamics of secondary combustion within the end-gas of SI engines. Nevertheless, it is important to acknowledge the limitations associated with the assumptions made in this study. These limitations are discussed in the following:

First, there is a lack of relevant experimental measurements concerning the flame and ignition characteristics of gasoline surrogates within the high-pressure and high-temperature combustion modes studied in this work. The chemical mechanisms utilized for PRF and PRF-E mixtures were validated in Publication I against flame speed and ignition delay times under conditions relevant to the initial state of the simulations. However, during the simulations, particularly the detonation propagation in Publication II and Publication III, both the pressure and temperature of the reaction zone exceeded the maximum validated range. Accordingly, it is of utmost importance to gain experimental data on gasoline surrogates
combustion within elevated temperatures and pressures.

Second, the computational setups used in this study involve simplified geometries and boundary conditions. The simulation cases investigated in the present work are initiated with simplified temperature stratification, homogeneous pressure, and mixture composition in 1D and 2D settings. However, it is important to note that the complex 3D flow field observed within the cylinder of SI engines, non-homogeneous mixture conditions, and the intricate effects of boundary conditions can all potentially influence the reactivity stratification and, consequently, the combustion dynamics within the end-gas of SI engines. Therefore, underlying 3D phenomena, boundary conditions, and non-homogeneous mixture composition may affect the reported observations.

Third, the present simulations primarily concentrate on the relatively large scales of the flow field and reaction front structure. In particular, in detonations resolving fine-scale turbulent dissipation may demand approximately $10^3$ times more grid points per induction length [70]. Current computational capabilities are unable to handle such high computational costs. In real scenarios, transient turbulence effects may indeed introduce variations in the observed combustion dynamics within the end-gas of SI engines.

Finally, despite the application of simulation efficiency approaches, the 2D simulations of hotspot ignition were limited to a small number of cases (in contrast to the hundreds of 1D simulations in Publication I and Publication II). These simulations were restricted only to the PRF mixture. This limitation was primarily due to the substantial computational cost associated with finite rate chemistry, which has remained the bottleneck in the investigation presented in this dissertation. Consequently, a more comprehensive examination of the effect of fuel chemistry within the 2D framework would necessitate higher computational capabilities or more efficient computational methods.

4.3 Future work

Based on the conclusions, limitations, and discussions provided in previous sections of this dissertation, the following suggestions for future work can be outlined:

1. Develop and validate simplified chemical mechanisms, along with their corresponding thermodynamic properties, for various gasoline surrogates under elevated pressures and low to intermediate temperatures that are relevant to the operational conditions of modern SI engines. The primary objective is to deliver CFD-friendly and precise chemical kinetics suitable for simulations that resemble conditions of knocking in SI engines.

2. Extend the studies in the present dissertation to gasoline surrogates
Conclusions and perspectives

with different components, including Toluene, as well as renewable oxygenates such as 2,5-dimethylfuran (DMF) and dimethyl ether (DME). The objective is to utilize the results and the 1D frameworks provided in Publication I and Publication II for gasoline surrogate mixtures with less known combustion characteristics, and subsequently compare the outcomes to those obtained from the examined PRF and PRF-E mixtures.

3. Explore the potential direct influence of low/intermediate heat release on the transition of combustion modes within the SI engine end-gas. Conducting reaction sensitivity analysis on ZND and 1D/2D transient detonation structures. The goal is to determine the effect of low/intermediate temperature ignition and the associated reactions in gasoline surrogates on the dynamics of secondary combustion events occurring within the SI engine end-gas.

4. Evaluate the performance of two different balancing strategies, OF-DLB and $DLB_{foam}$, in simulating reacting flows within the OpenFOAM framework. Additionally, explore potential enhancements and optimizations for the simulation efficiency methods within the $ARCFoam$ framework. The aim is to identify the most efficient combination of various numerical techniques that can improve simulations of high-speed reacting flows, particularly those relevant to knocking in the context of SI engines.

5. Incorporate the impact of more intricate boundary conditions, non-uniform mixture composition, and transient turbulence. The primary goal is to replicate the intricate flow patterns and mixture variations occurring within the cylinder in realistic SI engine combustion scenarios.
Appendix
The subsequent section presents an overview of the primary algorithms inherent to the AMR mesh balancer. As elucidated in section 2.3.2, the design of the mesh balancer is guided by two core objectives: firstly, leveraging the OF-DLB framework [92]; and secondly, preserving the integrity of the OF-AMR "tree" data structure (refinementHistory) on each processor [89]. Consequently, the term "AMR load balancer" characterizes the developed routines that synergistically combine the dual functionalities inherent in OpenFOAM namely AMR and load balancing. For detailed insights into the implementation of these codes, interested readers are directed to the GitHub repositories of OpenFOAM 10 [92] (load balancing) and BlastFOAM [25] (2D AMR).

Furthermore, this section addresses a solution to an issue observed in simulations of Publication III where simulation inefficiency stemmed from an escalating cell count during the AMR process. When simulating unsteady expanding or progressive phenomena using AMR, certain scenarios can lead to a substantial increase in the final cell count within the computational domain compared to the initial count. Such simulations, when initiated with a high number of processors designed to accommodate the eventual high cell count, can exhibit noticeable deficiencies during the initial stages due to the augmented number of processor boundaries and consequent inter-processor communication [12]. This necessitates domain "re-decomposition" to accommodate the newly generated cells via AMR, by increasing the number of CPUs as the simulation progresses.

### A.1 Creating coarse cell maps

The balance() function is invoked at the end of the AMR process in Foam::fvMeshTopoChangers::refiner::update() function. The new weights, indexing, and point maps are calculated based on the integrity of the parent cells on each processor. And in the end, are stored on the object registry. This treatment based on the Voskuilen [89] solution ensures that the "tree" data
structure (refinementHistory) is complete on each processor during the next step which is the cell distribution.

```cpp
void Foam::fvMeshTopoChangers::adaptiveFvMesh::balance ()
{

    // The following procedure is based on Voskuilen's
    // mesh Balancing solution for the original code, please
    // refer to:
    // https://github.com/tgvoskuilen/meshBalancing/blob/
    // meshBalancing-.3.x/dynamicRefineBalancedFvMesh/
    // dynamicRefineBalancedFvMesh.C

    const labelIOList& cellLevel = meshCutter().cellLevel();
    Map<label> coarseIDmap(100);
    labelList uniqueIndex(mesh().nCells(),0);
    label nCoarse = 0;

    forAll(mesh().cells(), cellI)
    {
        if
        {
            cellLevel[cellI] > 0
            && meshCutter().history().visibleCells()[cellI] >= 0
        }
        {
            label parentI = meshCutter().history().parentIndex(cellI);

            if (parentI >= 0)
            {
                uniqueIndex[cellI] = mesh().nCells() + topParentID
                (             
                    meshCutter().history().parentIndex(cellI)
                );
            }
            else
            {
                uniqueIndex[cellI] = cellI;
            }
        }
        else
        {
            uniqueIndex[cellI] = cellI;
        }
    }
}
```
if ( coarseIDmap.insert(uniqueIndex[cellI], nCoarse) )
{
    ++nCoarse;
}

labelList localIndex(mesh().nCells(),0);
pointField coarsePoints(nCoarse,vector::zero);
scalarField coarseWeights(nCoarse,0.0);
label nRefinementDimensions(mesh().nGeometricD());

forAll(uniqueIndex, cellI)
{
    localIndex[cellI] = coarseIDmap[uniqueIndex[cellI]];

    // nRefinementDimensions is the general
    // number of refinement dimensions, i.e. 1D, 2D or 3D.

    label w = (1 << (nRefinementDimensions*cellLevel[cellI]));

    coarseWeights[localIndex[cellI]] += 1;
    coarsePoints[localIndex[cellI]] += mesh().C()[cellI]/w;
}

// End Voskuilen’s method
// storing the values into ObjectRegistered variables

PPpoints = coarsePoints;
lI = localIndex;
cW = coarseWeights;

A.2 AMR mesh balancing

The update() function of the cell distributor is invoked following the execution of AMR, specifically after the refiner::update() function. Subsequently, an imbalance calculation is performed on each processor in
the new adaptiveLoadBalancer class, which assesses the deficit between the processor cell count and the ideal average number of cells per processor. If the computed imbalance surpasses the user-defined maxImbalance threshold, a new balanced cell distribution is generated by invoking the distribute(distribution) function. This process employs either the zoltan [15] or scotch [10] algorithms. Subsequent to this process, the geometry associated with the new cell distribution is reconstructed on each processor, and the associated fields are redistributed between the processors.

```cpp
bool Foam::fvMeshDistributors::adaptiveLoadBalancer::update()
{
    const fvMesh& mesh = this->mesh();
    bool redistributed = false;

    if
    {
        Pstream::nProcs() > 1
        && mesh.time().timeIndex() > 1
        && timeIndex_ != mesh.time().timeIndex()
    }
    {
        timeIndex_ = mesh.time().timeIndex();
        const scalar timeStepCpuTime = cpuTime_.cpuTimeIncrement();

        // CPU loads per cell
        HashTable<cpuLoad*> cpuLoads(this->mesh().lookupClass<cpuLoad>());

        if (mesh.time().timeIndex() % redistributionInterval_ == 0)
        {

            pointField PPpoints;
            labelList localIndex;
            scalarField coarseWeights;

            if
            {
                mesh.objectRegistry::
                    foundObject<pointIOField>("PPpoints")
                && mesh.objectRegistry::
                    foundObject<scalarIOField>("coarseWeights")
```
&& mesh.objectRegistry::
    foundObject<labelIOList>("localIndex")

{

    PPoints =
        mesh.lookupObject<pointIOField>("PPpoints");
    localIndex =
        mesh.lookupObject<labelIOList>("localIndex");
    coarseWeights =
        mesh.lookupObject<scalarIOField>("coarseWeights");

}

// Mesh related load

label nGlobalCells =
    mesh.globalData().nTotalCells();
scalar idealNCells =
    scalar(nGlobalCells)/scalar(Pstream::nProcs());
scalar localImbalance =
    mag(scalar(mesh.nCells()) - idealNCells);

Foam::reduce(localImbalance, maxOp<scalar>());
scalar imbalance = localImbalance/idealNCells;

if (imbalance > maxImbalance_)
{
    Info<< "Redistributing mesh with imbalance:\n"    << imbalance << endl;

    const labelList distribution
    {

        distributor_->decompose
        (        
            mesh,
            localIndex,
            PPoints,
            coarseWeights
        )
    };

    distribute(distribution);
A.3 redecomposePar Utility

The redecomposePar utility functions similarly to decomposePar, which facilitates domain decomposition before the simulation. However, this utility employs a modified decomposer (cell distributor) that operates based on the complete tree data structure (refinementHistory), i.e. Voskuilen’s solution [89], during the cell map generation phase. It is important to note that only automatic mesh distribution algorithms, specifically the zoltan [15] or scotch [10] algorithms, are compatible with the redecomposePar utility. To achieve this, the distributeCells() function from the domainDecomposition class needs to be modified as shown below:

```cpp
Foam::labelList Foam::domainReDecomposition::distributeCells()
{
    Info<< "Calculating distribution of cells" << endl;

    cpuTime decompositionTime;
    labelList result;

    IOobject io
    {
        "refinementHistory",
        completeMesh().facesInstance(),
        polyMesh::meshSubDir,
        completeMesh(),
        IOobject::READ_IF_PRESENT,
        IOobject::NO_WRITE
    };
```
typeIOobject<hexRefRefinementHistory> rio (io);
rio.rename("refinementHistory");

bool haveFile = returnReduce
{
    rio.headerOk(),
    orOp<bool>()
};

if (haveFile)
{
    hexRefRefinementHistory refHistoryPtr_(io);
    labelIOList cellLevel
    {
        IOobject
        {
            "cellLevel",
            completeMesh().facesInstance(),
            polyMesh::meshSubDir,
            completeMesh(),
            IOobject::READ_IF_PRESENT,
            IOobject::AUTO_WRITE
        },
        labelList(completeMesh().nCells(), 0)
    };

    // The following procedure is based on Voskuilen’s
    // mesh Balancing solution for the original code, please
    // refer to:
    // https://github.com/tgvoskuilen/meshBalancing/blob/
    // meshBalancing-.3.x/dynamicRefineBalancedFvMesh/
    // dynamicRefineBalancedFvMesh.C

    Map<label> coarseIDmap(100);
    labelList uniqueIndex(completeMesh().nCells(), 0);

    label nCoarse = 0;

    forAll(completeMesh().cells(), cellI)
    {
        if
        {
            cellLevel[cellI] > 0
            && refHistoryPtr_.visibleCells()[cellI] >= 0
        }
{
    label parentI = refHistoryPtr_.parentIndex(cellI);

    if (parentI >= 0)
    {
        uniqueIndex[cellI] =
            completeMesh().nCells() + topParentID
            (refHistoryPtr_.parentIndex(cellI),
                refHistoryPtr_)
    }
    else
    {
        uniqueIndex[cellI] = cellI;
    }
}
else
{
    uniqueIndex[cellI] = cellI;
}
"if ( coarseIDmap.insert(uniqueIndex[cellI], nCoarse) )
{
    ++nCoarse;
}

labelList localIndex(completeMesh().nCells(),0);
pointField coarsePoints(nCoarse,vector::zero);
scalarField coarseWeights(nCoarse,0.0);
label nRefinementDimensions(completeMesh().nGeometricD());

forAll(uniqueIndex, cellI)
{
    localIndex[cellI] = coarseIDmap[uniqueIndex[cellI]];

    label w = (1 << (nRefinementDimensions*cellLevel[cellI]));
    coarseWeights[localIndex[cellI]] += 1;
    coarsePoints[localIndex[cellI]] += completeMesh().C()[cellI]/w;
}
// End Voskuilen's method

const dictionary decomposeParDict =
    decompositionMethod::decomposeParDict(runTimes_.completeTime());

result =
    decompositionMethod::NewDecomposer(decomposeParDict)->decompose
    (completeMesh(),
     localIndex,
     coarsePoints,
     coarseWeights);

Info<< "\nFinished decomposition in"
    << decompositionTime.elapsedCpuTime()
    << "s" << endl;

return result;
} else {

FatalErrorInFunction
    << "No refinementHistory was found."
    << exit(FatalError);

}
}
References


References


References


References


Improving efficiency and reducing emissions in internal combustion engines is crucial for achieving emission goals. However, the optimization process is significantly challenged by the occurrence of knocking phenomena. Understanding the complex relationship between knocking and various factors, such as fuel chemistry, pressure oscillations, and auto-ignition front propagation, is still in its early stages. To address these aspects, this dissertation focuses on the thermophysical processes responsible for abnormal combustion under conditions relevant to SI engines. By exploring the underlying thermophysical mechanisms, we aim to elucidate the details of combustion and pressure wave generation processes of knock in modern engines. Notably, a systematic theoretical/numerical approach and an efficient computational fluid dynamics (CFD) solver were proposed to address the underlying thermophysical events within the end gas of SI engines. The simulation results shed light on the influence of negative temperature coefficient (NTC) chemistry on the appearance of different ignition regimes, propagation patterns, and associated variations in the flow field within the end gas.