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de thèse. Le lecteur pourra sentir l'empreinte de Thierry, avec son recul et son expérience, dans les différentes parties de ce manuscrit. Aussi, l'oeil aiguisé et le tact absent de Thierry vous rappellent très souvent qu'il est inutile d'essayer de tromper le chef. Je suis bien obligé aujourd'hui de t'avouer, pauvre thésard contraint de lire ce manuscrit, que j'ai bien essayé. Mes tentatives sont toutefois restées vaines. Ensuite, il y a Olivier que je ne remercierai jamais assez pour ses différents conseils et son impact monstre dans le travail présenté dans ce manuscrit. Parler d'Olivier est certainement la partie la plus difficile de ces remerciements. Comment résumer ce que je pense de ce grand Monsieur en quelques lignes? Est-ce seulement possible? Ce sont les questions qui me taraudent l'esprit à l'heure où j'écris ce paragraphe. Laissant ça pour une autre occasion, je me permets une simple révérence: Chapeau bas l'artiste!

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Un homme sérieux a peu d'idées. Un homme à idées n'est jamais sérieux.

Montesquieu, 1689-1755

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Part I

General concepts of self-propagating combustion waves

Chapter 1

Introduction

1.1 Explosion hazards

Every year, mining, process and energy industries suffer billions of dollars of losses worldwide due to gas explosions. In addition to the direct damage to the industrial plants, these explosions often lead to long business interruptions, causing secondary costs that need to be considered. Indeed present-days production chains exhibit a high degree of consolidation, which, in case of explosion, can lead to catastrophic financial losses. Moreover, explosion accidents are often tragic and lead to a high number of severe injuries and fatalities. Many severe industrial disasters have been reported in the past years:

- April 14th, 1944, India Cargo explosion in Mumbai : 1300 fatalities, 3000 injuries;
- July 11th, 1978, Spain Tanker road accident: 217 fatalities, 200 injuries;
- November 19th, 1984, Mexico Series of massive explosions in a liquid petroleum gas tank in San Juanico: 500 fatalities, 6000 injuries;
- March 23rd, 2005, US Explosions in a BP Texas refinery: 15 fatalities, 180 injuries;
- December 11th, 2005, England Buncefield disaster: 43 injuries;
- April 20th, 2010, Golf of Mexico Explosion in Off-shore petroleum platform: 11 fatalities;
- August 25th, 2012, Venezuela Explosion in gas refinery in Amuay: 48 fatalities;
- August 12th, 2015, China Series of massive explosions at a container storage station in the Port of Tianjin: 173 fatalities, 797 injuries.

This non exhaustive list of disasters shows that large explosion hazards can have unforeseeable social and political impacts that can exceed the direct economical damages. Sound knowledge of explosions physics is therefore of vital importance for the prediction of these extreme scenarios.

First, the term "explosion" needs to be defined. It refers to a violent event able to generate considerable overpressure. An insightful definition was proposed by F. M. Global (2013): "an explosion is a rapid transformation of potential physical or chemical energy into mechanical energy and involves the violent expansion of gases". In this definition, the



Figure 1.1: Position of the Vapour Cloud Explosions (VCE) in the classification of explosions in chemical process industries proposed by Abbasi et al. (2010).

terms "physical" and "chemical" refer to the way the energy, that enabled the explosion, is accumulated:

- Physical explosions are caused by the sudden release of an energy accumulated by gas or volatile liquid heating in a containment for example, or by simple gas compression, generating overpressure. The release can be the result of the failure of the containment.
- Chemical explosions are due to the release of energy by chemical reactions causing pressure build-up. Chemical explosions can be either homogeneous or reaction front driven.

Independently of their nature, explosions generate overpressure. The level of pressure produced during an explosion is the critical parameter that will determine the severity of the damages inflicted on buildings and civilians. Because the losses vary considerably from an explosion to another, it seems that some mechanisms can greatly increase the levels of overpressure generated. Identifying and understanding these mechanisms is crucial to provide engineers with guidelines for implementing preventive and mitigative measures.

Abbasi et al. (2010) proposed different subclassifications of explosions in process industries. A part of this classification is shown in Fig. 1.1. The mechanisms involved vary considerably from an explosion to another. This thesis focuses on reaction front driven explosions and more specifically on Vapour Cloud Explosions (VCE). A sequence of events commonly involved in VCEs is displayed in Fig. 1.2. Reactive explosions occur when a cloud of premixed reactive mixture has enough time to form after a gas leak. An external source of energy E_s deposited over a duration τ_s can then ignite the mixture and a reaction wave emerges from the source point, consuming the reactants ahead of it to form products at its tail. The origin of this external energy is generally attributed to sparks, walls heating, annex explosions etc... Generally speaking, two distinct types of self-propagating reaction waves can emerge from the region where energy is deposited (their structure is shown in Fig. 1.3):

• Premixed flames are very subsonic and quasi-isobaric combustion waves. Their



Figure 1.2: Commonly observed event tree leading to gas explosions in buildings.

propagation is governed by thermal conduction as well as by chemical reaction. The temperature inside the flame front gradually increases due to, first, diffusive flux of energy by conduction, before exothermic reactions increase the temperature further to reach the equilibrium temperature of the products. Premixed flames are initiated if E_s is relatively low but sufficient to activate chemical reactions. The term "deflagration" generally refers to premixed flames that have reached high propagation velocity with respect to a fixed point.

• Detonations are supersonic waves. Their propagation is governed by shock dynamics. Contrary to premixed flames, the reaction zone is not initiated by heat diffusion. Instead a strong shock brings the reactants to the Von-Neumann state (VN) where the temperature is high enough to initiate reactions. Detonations can be directly initiated if E_s is sufficiently high and deposited on a very low time scale τ_s . However, this is not a necessary condition and detonations can be triggered by another mechanism involving the spontaneous ignition of a pocket of fresh gases provided some precondition is met.

Because of the presence of a strong shock coupled to a reaction zone, detonations are fare more destructive than deflagrations. Therefore, the initiation of the former waves has to be carefully studied to identify the possible mechanisms that can trigger this escalation in the explosion hazard. Generally speaking, detonations are observed when: 1) a strong blast forms and decays to a self-propagating detonation front; 2) a premixed flame forms and creates, during its propagation, the ideal conditions for the coupling between a shock and a reaction wave, thereby giving rise to a detonation. Both scenarios are discussed briefly in Sections 1.2 and 1.3.

1.2 Direct detonation initiation

A detonation wave can be initiated far away from the energy deposition locus if sufficient energy E_s is deposited (quasi-)instantaneously (Clavin and Searby (2016); Lee (2008)).



Figure 1.3: The inner structure of premixed flames and detonations. In a premixed flame (left), reactions are initiated by the gradual increase of temperature due to the diffusive flux of heat. The reaction zone, in a detonation (right), is initiated by the abrupt temperature increase induced by the leading shock. The indices u and b indicate the fresh and burnt gases respectively. The bold arrow indicates the direction of propagation.

 E_s must then be higher than a certain critical value $E_{s,1}^c$ (see Fig. 1.4(a)). In this case, a strong blast wave is formed and decays to a self-propagating detonation wave at a certain distance r_d from the ignition source (see Fig. 1.4(b)). Once the detonation regime is reached, a quasi-steady high overpressure level is observed. Note that r_d depends on E_s . On the other hand, no detonation is observed if E_s is just sufficiently high to initiate a blast wave (i.e. $E_s > E_{s,2}^c$) but lower than the critical value $E_{s,1}^c$. The blast simply decays to a relatively weak shock. Note that the damages inflicted on buildings and civilians can be extreme in these cases because of the shock generated initially.

In terms of safety, it is important to predict the critical energy deposition $E_{s,1}^c$ that can lead to detonation onset. This has been assessed both theoretically (He and Clavin (1994)) and experimentally (Bach et al. (1969); Lee (1984)). $E_{s,1}^c$ was found to be quite large and can explain the formation of gaseous detonations in free space. In this thesis, we are interested in semi-confined and confined configurations, in which direct detonation initiation is not the commonly observed scenario. There is, however, a mechanism by which a detonation can still form far away from a mild ignition source if certain conditions are met. This is discussed briefly in the next section.

1.3 Delayed detonation initiation

This thesis focuses on explosions occurring on configurations that are either semi-confined or confined. In such configurations, the explosion is generally initiated by a mild ignition, i.e. $E_s \ll E_{s,2}^c$. A deflagration front then emerges from the ignition source. Since a deflagration wave corresponds to a subsonic regime of propagation, there must exist an



Figure 1.4: (a) Depending on the energy deposited E_s to ignite the mixture, multiple scenarios can arise. This thesis addresses the case of a mild ignition that leads to deflagration initiation. (b) Typical maximum overpressure (overP) evolution with the distance r_s from the ignition source. When the energy input to the system is sufficient, i.e. $E_s > E_{s,1}^c$, a blast wave transitions to a detonation wave with a quasi-steady high overpressure level. Conversely, if $E_{s,2}^c < E_s < E_{s,1}^c$, the blast wave decays to form a relatively weak shock.

acceleration process that leads to the fast flames observed during explosions. A "strong" acceleration stage is a necessary condition before a detonation initiation can be observed.

1.3.1 Flame acceleration: from very subsonic to very fast flames

As discussed above, a VCE can be defined as a "rapid" release of chemical energy. Given that premixed flames are subsonic combustion waves, one may ask where the term "rapid" comes from, in the case of deflagrations. The answer lies in the ingredients controlling the flame propagation speed: 1) flames consume the reactants at their vicinity at velocity $\mathcal{V}_{consumption}$ that depends on the local conditions in the fresh gases ($\mathcal{V}_{consumption}$ is also the velocity of the flame with respect to the fresh gases); 2) similarly to acoustic waves, flames are also advected by the flow at speed \mathcal{V}_{flow} . With respect to a fixed point, the flame propagation speed \mathcal{V}_{flame} is then given by the relation: $\mathcal{V}_{flame} = \mathcal{V}_{consumption} + \mathcal{V}_{flow}$. An important feature of self-propagating flames is that $\mathcal{V}_{consumption}$ can be influenced by numerous mechanisms, described in Chapter 3, which makes these reaction waves intrinsically unstable (Lee (2008)). A constructive feedback between the unsteady flame and the flow ahead of it may give rise to a Flame Acceleration (FA) process responsible for the emergence of the fast flames observed in real explosions.

As the flame accelerates, the exothermal reactions inside the deflagration front release more energy leading to more pressure build-up. A typical FA scenario is displayed in Fig. 1.5(a). The maximum overpressure, and consequently the degree of damages observed during the gas explosion, depends on the final level of acceleration reached by the deflagration and varies considerably from a configuration to another.

In order to predict the maximum overpressure reached during FA in VCEs, empirical methods are still used nowadays to design industrial plants. A compilation of such formulas is proposed in Bradley and Mitcheson (1978) and Molkov et al. (1997). However, their applicability is limited to cases identical (or very close) to the setup on which such



Figure 1.5: (a) The solid line represents a typical flame acceleration scenario that leads to high damages. The bold line corresponds to the case where DDT is observed with a drastic increase in the damages inflicted. (b) A deviation from the typical FA scenario is observed if mitigative procedures are applied. A good mitigative procedure for VCE does not result in flame extinction, instead the flame must continue to burn reactive gases but with minimum damage.

correlations have been built. Indeed, FA is controlled by instabilities and as such is highly sensitive to initial and boundary conditions (in addition to other parameters discussed later in the thesis). Such correlations can hardly take into account the complexity of the industrial structures and a deviation of the observed overpressure from the one predicted with such correlations is often observed.

1.3.2 Deflagration to detonation transition

Flame acceleration results in very fast deflagrations, sometimes supersonic with respect to the laboratory frame. Once this regime is reached, the appropriate conditions for detonation onset may be reached as displayed in Fig. 1.5(a). This transition is called the Deflagration to Detonation Transition (DDT). A variety of events/ingredients have been shown, theoretically, experimentally and numerically, to trigger DDT under specific flow conditions. The challenge is to understand the interplay among these ingredients and the relative importance of each one.

DDT marks a brutal escalation in the explosion hazard. To implement preventive measures against DDT, multiple criteria, summarized in Ciccarelli and Dorofeev (2008), have been proposed. Assuming that a deflagration has already accelerated to a certain critical supersonic velocity that depends on the configuration, it is possible to define a criterion for the onset of detonation. The problem is that boundary conditions play an important role in both the onset and the propagation of detonations, so that it is only meaningful to discuss possibility of DDT for specific boundary conditions (see Lee (2008) and Chapter 8 of the manuscript). Therefore, such criteria can hardly be of relevance to complex industrial plants.

1.4 Including DDT in risk assessment: past accidents

Large scale experiments, mentioned in Ciccarelli and Dorofeev (2008) and Lee (2008), provide ample evidence that DDT can occur in a VCE. Not only that, the detonation, once initiated, can continue through unobstructed regions of the cloud, thereby inflicting damage to areas far away from the onset locus. The experimental evidence points out the necessity to include the DDT phenomenon in risk assessment, despite its low probability of occurence. Recent industrial accidents show that a simple tank switch failure can trigger a cascade of events leading to huge industrial disasters:

- Buncefield, UK, 2005: a tank, filled with unleaded petrol, began to overflow. This was attributed to the failure of a switch, which would have detected that the tank was full and shut off the supply. This resulted in the rapid formation of a rich fuel and air vapour. By the time the first explosion occurred, the cloud had spread beyond the boundaries of the site. Therefore, the devastated area exceeded the industrial site. The investigation report, issued by the british Government in 2008, pointed out some indicators of DDT. In particular, the damages recorded on areas that do not present signs of congestion (see Fig. 1.6 (top)) suggest the self-propagation of a detonation front. The disaster did not result in any fatality but it caused over 200 million euros of financial losses.
- Jaipur, India, 2009: a vapour petrol leak was detected but the non-observance of (normal) safety procedures led to a strong explosion. The evidence of severe pressure damage can be observed throughout the area, as illustrated in Fig. 1.6 (bottom). The level of damage observed indicates pressures of several bars and is consistent with the damage caused by the passage of a detonation. 11 people were killed in the disaster and the financial losses add up to nearly 40 million euros.

These severe accidents demonstrate the necessity to implement measures than can either prevent or reduce the impact of explosions. In the last case, this procedure is called mitigative.

1.5 From preventive to mitigative measures: flame inhibition

For safety engineers, there are two possible ways to deal with VCE's:

• **Preventive measures.** The problem with these measures is that their effectiveness can hardly be always guaranteed. First, a large spectrum of mechanisms can lead to strong FA. Preventive measures may limit the impact of some mechanisms that are dominant under certain conditions. But they can fail when some unexpected scenarios give rise to other processes that can trigger a cascade of dramatic events as often highlighted in industrial disasters. Second, even though criteria exist for the detonation propagation limit beyond which the self-sustained propagation of a detonation wave is not possible, they are strongly dependent on the boundary conditions. Therefore, one can even question the universality of such preventive



Figure 1.6: Damages recorded during the Buncefield (top) and the Jaipur (bottom) incidents. There are some indicators of DDT during these disasters.

measures when taking into account the complexity of the explosion scenario in real industrial plants.

• Mitigative measures. Since the first explosion accidents, it has become clear that preventive guidelines must be supported by mitigative procedures that can activate when a gas leak or an ignition is detected. Water sprays are an example of such measures. Their applicability to vapor cloud explosions has been shown by a number of authors (Acton et al. (1991); Bjerketvedt et al. (1997); Van Wingerden (2000)). Water droplets act as a heat sink and their effectiveness depends on their size. Water sprays are a promising mean for explosion mitigation since they are relatively cheap and most industrial installations are already equipped with a fire fighting system via which water can be delivered. However, there are also a number of drawbacks. The main one, identified by practical studies performed on the petrochemical complex of Samsung-Total Petrochemicals in South Korea (Hoorelbeke (2011)), is that the quantities of water needed to strongly reduce the overpressure may be huge.

For the case of VCE's however, there is a condition that a mitigative measure must verify before it can be considered as an operational system: the cloud of reactive mixture that have formed due to the gas leak must be eliminated. This means that any procedure that can eventually result in flame extinction can not be applied to VCE's, as highlighted in Fig. 1.5(b). Indeed, if at some point the flame is suppressed, a second explosion can still occur later in the remaining reactive cloud. Because of the high temperatures and pressures created by the first explosion, the second ignition may lead to higher damage. Therefore, an effective mitigative measure for VCE's is one that strongly reduces the overpressure without extinguishing the flame. Because VCE's are reaction front driven, an efficient way to achieve this is to reduce the overall exothermic reaction rate in the flame, so that only very weak FA is achieved during the deflagration propagation. This mechanism is referred to, by Linteris (2004), as **flame inhibition** and will be discussed in more details in Part III of the manuscript. It can be achieved, for example, by injection of a powder ahead of the flame, as studied in this PhD.

1.6 The role of Safety Computational Fluid Dynamics (SCFD)

As explained in the previous section, VCE scenarios are complex and controlled by various mechanisms. The interplay among them is not yet entirely understood. The same remark applies to mitigative measures. The mechanism by which the latter act on the explosion has to be characterized in details, in order to avoid surprising counter-effects. Understanding all these intricate processes is of vital importance and requires detailed experimental diagnostics. Coupling accurate numerical simulations to well documented experiments can allow an elaborate description of these phenomena.

Considered too expensive a dozen years ago, CFD is now more and more included in risk assessment studies. The main advantage of CFD, compared to phenomenological and empirical methods, is to allow access to flow and flame structures. This is crucial to understand the mechanisms involved in VCE. The main limit to the use of SCFD is the complexity of the configurations considered. Industrial plants are often complex, including large scale (buildings, tanks, etc...) to small scale (pipes, instrumentation devices, etc.) structures. The same remark holds for the time scales (chemistry, instabilities, detonation, flames, shocks, etc.). The spectrum of time and length scales is large and implies huge computational resources, that may not even be available.

In order to reduce the computational cost of such simulations, modeling the entire turbulent spectrum is a usual approach known as Unsteady Reynolds Average Navier-Stokes (URANS). It has been adopted by most of the licensed SCFD codes. A series of benchmark tests have been conducted Baraldi et al. (2009); Garcia et al. (2010); Makarov et al. (2010) showing acceptable agreement with experimental data. It is however important to note that most of these simulations have been conducted with an a priori knowledge of the experimental results, allowing the user to tune the model coefficients to match the desired curves. According to Pope (2001), the most widely used RANS turbulent models $(k - \varepsilon$ Jones and Launder (1972); Launder and Sharma (1974), $k - \omega$ Wilcox (2008)) can fail profoundly when applied to inhomogeneous turbulent flows. Even in the case of Reynolds-stress models (Pope (2001)) a calibration of the constants is necessary for many flows. The relevance of such models to transient phenomena like VCE is therefore doubtful. As indicated by Ciccarelli and Dorofeev (2008), "RANS codes are missing the essential physics that are responsible for flame acceleration and DDT and therefore can only be used for qualitative information". Blind simulations of realistic explosion scenarios using these codes are still to be performed to assess their capacity to correctly predict the experimental observations.

With the increase of the computational power of present-days computers, the Large Eddy Simulation (LES) approach emerges as a promising alternative to URANS. In LES, the large flow structures are resolved explicitly whereas small structures are modeled. Many authors (Makarov and Molkov (2004); Molkov et al. (2007); Gubba et al. (2008, 2009); Zbikowski et al. (2008); Ibrahim et al. (2009); Gubba et al. (2011); Wen et al. (2012); Quillatre et al. (2013); Xu et al. (2015); Zhao et al. (2017); Tolias et al. (2017); Volpiani et al. (2017a); Vermorel et al. (2017)) have shown the capacity of LES to capture the transient effects related to turbulence and flame-turbulence interactions during deflagra-



Figure 1.7: Typical vapour cloud explosion scenarios addressed in this thesis.

tions in small-scale closed and vented chambers. However, LES shows serious limitations when used to compute deflagrations in large scale configurations ($550m^3$ in the study of Molkov and Makarov (2006)). In such domains, the grid resolution is degraded to keep the computational cost reasonable, which reduces considerably the part of the physics resolved. One may even question the LES designation in these cases.

While small scale deflagrations have been computed with good accuracy using LES, DDT has mostly been simulated with Direct Numerical Simulations (DNS) (see the work of Oran and coworkers summarized in Oran and Gamezo (2007)). The reason behind this is that very small grid cells are needed to capture both the flow, the flame and the detonation structures. This reduces considerably the dimensions of the problem that can be considered. Even for "small" domains, three dimensional DNS is still unaffordable nowadays and in most cases only two-dimensional domains are considered. The 2D-DNS aims at highlighting important mechanisms that can trigger DDT.

1.7 Objective of the thesis

The objective of this thesis is to investigate numerically reaction front driven explosions. First, conservation equations for reactive flows and some modeling aspects are recalled in Chapter 2. Some theoretical aspects of premixed flames and detonations are discussed in Chapters 3 and 4 respectively. FA, DDT and flame inhibition are investigated in this thesis, each problem requiring a specific computational strategy. For the sake of clarity, the different methods considered are summarized in Chapter 5 and a justification is provided whenever a change in strategy was deemed necessary. Parts II and III of the manuscript are dedicated to the presentation of the results.

As displayed in Fig. 1.4, all the scenarios considered in this work start with a mild ignition, from which a subsonic deflagration front emerges. This manuscript tries to answer two major questions: 1) What are the mechanisms responsible for the extreme damages observed during VCE's? 2) What is the underlying mechanism behind some efficient mitigative measures?

Part II: What are the mechanisms responsible for the extreme damages observed during VCE's?

To answer this question, the scenario leading to DDT is investigated in two steps: a case resulting in fast deflagrations followed by a case where the transition is observed. The objective is to identify the necessary conditions that the flame acceleration phase must satisfy to trigger DDT. As discussed earlier, boundary conditions play an important role during both FA and DDT. Therefore, the discussion can only be meaningful if the same configuration is used for both cases. The Gravent explosion channel (Vollmer et al. (2012)) offers a large database of experimental results tailored for this investigation. The selected experiment, presented in details in Chapter 6, considers a long obstructed channel filled with premixed hydrogen/air mixtures at various compositions. Both FA and DDT have been observed depending on the equivalence ratio ϕ of the mixture.

Part II.a: Flame acceleration, from subsonic to very fast deflagrations

First a case corresponding to a lean ($\phi = 0.52$) premixed hydrogen/air mixture is considered. The results are presented in Chapter 7. The crucial importance of repeated flame-obstacle interactions in producing very fast deflagrations is highlighted. The high levels of pressure recorded during the experiments are explained. The study ends with a theoretical study of the flame journey in a flow contraction to deepen our understanding of the flame/obstacle interaction.

Part II.b: Transition from deflagration to detonation phenomena

By increasing the equivalence ratio of the mixture from $\phi = 0.52$ to $\phi = 1$, DDT is observed in the experiments. To identify the possible mechanisms that can explain DDT in this case, it is mandatory to understand why no detonation onset occurred in the case $\phi = 0.52$ despite the strong shocks produced ahead of the flame (Chapter 8). This prepares the ground for the numerical investigation of the detonating case, presented in Chapter 9. Particular attention is drawn to the impact of the chemistry modeling on the detonation scenario.

Part III. What is the underlying mitigative mechanism behind some solid inhibitors?

Part II focuses on possible VCE scenarios when no mitigative measure is applied. As discussed in Section 1.5, it is possible to considerably reduce the pressure levels produced during these events using techniques that are suited for reactive front driven explosions. Chapter 10 introduces the general concept of the chemical flame inhibition and its applicability to gas explosions. A particular attention is drawn to the sodium bicarbonate, a very efficient solid inhibitor. Chapter 11 presents the different methods used to derive a model able to reproduce the first order effects of sodium bicarbonate particles on a self propagating methane/air flame with reasonable computational cost. This model is then employed to investigate numerically the flame/inhibitor powder interaction. The results are discussed in Chapter 12. First, Section 12.1 focuses on the impact of the particle size

and powder stratification on the flame/inhibition process. Then, in Section 12.2, threedimensional simulations of a methane/air deflagration inhibition by sodium bicarbonate powders are presented. The suitability of the model for three-dimensional simulation is demonstrated. Chapter 2

Conservation equations and models for reactive flows

2.1 Introduction

The objective of the thesis is to investigate different gas explosion scenarios using numerical simulations. To describe fluids, a set of equations is needed. The first proposition of such a system of equations describing inviscid three-dimensional flows is attributed to Leonhard Euler. The notion of viscosity was later included by Louis Navier and George Gabriel Stokes to form the present-days formulation. Navier-Stokes equations are a set of partial differential equations including highly non-linear terms which, in most cases, make finding analytical solutions almost insurmountable. The very few analytical solutions have been obtained in general by considering cases where the non-linear terms vanish naturally. In most cases however, Navier-Stokes equations must be dealt with numerically. This chapter introduces the different strategies employed to "resolve" these equations.

2.2 Multi-species reactive flows

Reactive flows involve multiple species reacting through a set of chemical reactions. To characterize a species k in a mixture of mass m, containing n moles and N species, the mass fractions Y_k and the mole fraction X_k are used:

$$Y_k = m_k/m \tag{2.1}$$

$$X_k = n_k/n \tag{2.2}$$

where m_k (n_k) is the mass (number of moles) of species k in a given volume and m (n) is the total mass (number of moles) in this volume respectively. By definition,

$$\sum_{k=1}^{N} Y_k = \sum_{k=1}^{N} X_k = 1$$
(2.3)

The mean molecular weight of the mixture M is defined using the molecular weight of each species W_k :

$$1/M = \sum_{k=1}^{N} Y_k / W_k \tag{2.4}$$

The heat capacities at constant pressure C_p and constant volume C_v are defined by:

$$C_{p} = \sum_{k=1}^{N} Y_{k} C_{p,k}$$
(2.5)

$$C_{v} = \sum_{k=1}^{N} Y_{k} C_{v,k}$$
(2.6)

where $C_{p,k}$ and $C_{v,k}$ are respectively the heat capacities at constant pressure and constant volume of species k.

2.3 Compressible multi-species reactive Navier-Stokes equations

Navier-Stokes equations include conservation equations for mass, momentum and energy, plus a conservation equation for each species considered in the mixture. When radiative source terms are neglected, these equations have the following form:

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_j} (\rho u_j) = 0 \tag{2.7}$$

$$\frac{\partial}{\partial t}(\rho u_i) + \frac{\partial}{\partial x_j}(\rho u_i u_j + p\delta_{i,j} - \tau_{i,j}) = 0$$
(2.8)

$$\frac{\partial}{\partial t}(\rho E) + \frac{\partial}{\partial x_j}(u_i(\rho E + p\delta_{i,j} - \tau_{i,j}) + q_j) = \dot{Q} + \dot{\omega}_T$$
(2.9)

$$\frac{\partial}{\partial t}(\rho Y_k) + \frac{\partial}{\partial x_j}(\rho Y_k u_j) = -\frac{\partial}{\partial x_j}(J_{j,k}) + \dot{\omega}_k \tag{2.10}$$

where the Einstein notational convention is used. ρ , $(u_i)_{i=1,3}$ and p are the density, the velocity and the static pressure respectively. \dot{Q} denotes an external energy source term that can be induced by a spark or a laser for example. In this thesis, ρ and p are related through the state equation: $\rho = rT/p$, where T is the temperature and r = R/M is the perfect gas constant $R = 8.3145 Jmol^{-1}K^{-1}$ divided by the mean molecular weight M. E is the sum of the sensible and kinetic energy.

Viscous and pressure tensors: $\tau_{i,j}$ and $-p\delta_{i,j}$

In both momentum (2.8) and energy (2.9) conservation equations, the term $\sigma_{i,j} = \tau_{i,j} - p\delta_{i,j}$ appears as the sum of the viscous tensor $\tau_{i,j}$ and the pressure tensor $-p\delta_{i,j}$. $\tau_{i,j}$ is defined by:

$$\tau_{i,j} = -\frac{2}{3}\mu \frac{\partial u_k}{\partial x_k} \delta_{i,j} + \mu \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right)$$
(2.11)

where μ is the dynamic viscosity. $\delta_{i,j}$ is the Kronecker symbol: $\delta_{i,j} = 1$ if i = j and 0 otherwise.

Diffusive flux of species $J_{i,k}$

In the species conservation equation (2.10), the diffusive flux of species $J_{i,k}$ appears in the right hand side. The exact evaluation of $J_{i,k}$ is complex since it involves species diffusion velocities which are obtained by solving a linear system of size N^2 in each direction of space, for each point and each time. Since mathematically this task can be difficult and costly (Ern and Giovangigli (1994)), an approximation is commonly used in most numerical codes. The J. O. Hirschfelder and Bird (1954) approximation replaces the binary mass diffusion coefficient $D_{k,j}$ of species k into species j by an equivalent diffusion coefficient of species k into the rest of the mixture D_k . This first order approximation however does not guarantee mass conservation and a correction velocity $(V_i^c)_{i=1,3}$ is introduced:

$$J_{i,k} = -\rho \left(D_k \frac{W_k}{M} \frac{\partial X_k}{\partial x_i} - Y_k V_i^c \right)$$
(2.12)

where

$$V_i^c = \sum_{k=1}^N D_k \frac{W_k}{M} \frac{\partial X_k}{\partial x_i}$$
(2.13)

Energy flux q_i

For a multi-species flow, the energy flux q_i is the sum of two terms: heat conduction and diffusion of species with different enthalpies.

$$q_i = -\lambda \frac{\partial T}{\partial x_i} - \rho \sum_{k=1}^N \left(D_k \frac{W_k}{M} \frac{\partial X_k}{\partial x_i} - Y_k V_i^c \right) h_{s,k}$$
(2.14)

where $h_{s,k}$ is the sensible enthalpy of species k and λ is the heat diffusion coefficient.

Species source terms

Consider a chemical system of N species reacting through M reactions:

$$\sum_{k=1}^{N} \nu'_{kj} \mathcal{M}_k \rightleftharpoons \sum_{k=1}^{N} \nu''_{kj} \mathcal{M}_k \quad \text{for} \quad j = 1, M$$
(2.15)

where \mathcal{M}_k is a symbol for a species k. ν'_{kj} and ν''_{kj} are the molar stoichiometric coefficients of species k in reaction j. The mass reaction rate $\dot{\omega}_k$ of species k is the sum of the rates $\dot{\omega}_{kj}$ produced by each reaction:

$$\dot{\omega}_{k} = \sum_{j=1}^{M} \dot{\omega}_{kj} = W_{k} \sum_{j=1}^{M} \nu_{k,j} \mathcal{Q}_{j}$$
(2.16)

where $\nu_{k,j} = \nu''_{kj} - \nu'_{kj}$ and Q_j is the progress rate of reaction j which is defined by:

$$Q_{j} = K_{f,j} \prod_{k=1}^{N} \left(\frac{\rho Y_{k}}{W_{k}} \right)^{\nu'_{kj}} - K_{r,j} \prod_{k=1}^{N} \left(\frac{\rho Y_{k}}{W_{k}} \right)^{\nu''_{kj}}$$
(2.17)

 $K_{f,j}$ and $K_{r,j}$ are the forward and reverse rates of reaction j and are usually modeled using the empirical Arrhenius law:

$$K_{f,j} = A_{fj} T^{\beta_j} \exp\left(-E_{a,j}/RT\right)$$
(2.18)

The reverse rates $K_{r,j}$ are computed from the forward rates through the equilibrium constants:

$$K_{r,j} = \frac{K_{f,j}}{\left(\frac{p_a}{RT}\right)^{\sum_{k=1}^{N} \nu_{k,j}} \exp\left(\frac{\Delta S_j^0}{R} - \frac{\Delta H_j^0}{RT}\right)}$$
(2.19)

where $p_a = 1bar$. ΔH_j^0 and ΔS_j^0 are respectively the enthalpy (sensible and chemical) and entropy changes for reaction j:

$$\Delta S_j^0 = \sum_{k=1}^N \nu_{k,j} W_k s_k(T)$$
(2.20)

$$\Delta H_j^0 = \sum_{k=1}^N \nu_{k,j} W_k(h_{s,k}(T) + \Delta h_{f,k}^0)$$
(2.21)

 $\Delta h_{f,k}^0$ is the enthalpy of formation of species k at temperature $T_0 = 0K$, and allows to defined the heat release due to combustion $\dot{\omega}_T$:

$$\dot{\omega}_T = -\sum_{k=1}^N \dot{\omega}_k \Delta h_{f,k}^0 \tag{2.22}$$

Finally the sensible enthalpy $h_{s,k} = \int_{T_0}^T C_{p,k} dT$ is defined to satisfy $h_{s,k}(T = T_0) = 0$. The sensible energy is then defined by: $e_{s,k} = \int_{T_0}^T C_{v,k} dT$ so that $e_{s,k} = h_{s,k} - RT/W_k$.

Transport properties

To evaluate transport properties at reasonable cost, other approximations are applied. First, the Prandtl number $Pr = \mu C_p / \lambda$, which compares the momentum and heat transport, is set constant. Second, the Schmidt number $Sc_k = \mu / (\rho D_k)$ of each species k, which compares momentum and species diffusion, is assumed constant as well. This leads to a simplified relation for the species diffusion coefficient, which can then be evaluated using the viscosity μ . In this thesis, the temperature dependence of μ is assumed to follow a power law:

$$\mu = \mu_{ref} \left(T/T_{ref} \right)^b \tag{2.23}$$

where μ_{ref} is the viscosity at $T = T_{ref}$, and b a constant of the model in the range [0.5, 1.]

2.4 Different approaches to kinetic modeling

The species chemical source terms $\dot{\omega}_k$ in the energy and species balance equations (2.9, 2.10) describe the species conversion rates during the combustion process. "Real" flames involve many reactive molecules with a large range of reaction time scales τ_r . Thanks to

collision theory and quantum mechanics codes, the rate k_r of chemical reactions can be evaluated. In general, an Arrhenius law is used to get the expression of k_r :

$$k_r \approx A \exp(-E_a/RT) \tag{2.24}$$

This law basically states that the energy brought by colliding reactive molecules has to be higher than a certain energy E_a to open bonds and initiate reaction. Furthermore, the colliding molecules must be oriented in a manner favorable to the necessary rearrangement of atoms and electrons. The frequency of collisions in the correct orientation is given by A. To take into account the dependence of this collision frequency on temperature, A exhibits usually the form: $A \equiv AT^{\beta}$. It is important to note that the Arrhenius formulation is a drastic assumption of the intricate molecular collision phenomena. We present in the following a brief overview of different approaches to chemistry description that are based on an explicit chemical scheme. Tabulation methods (Maas and Pope (1992); Gicquel et al. (2000); van Oijen et al. (2001); Fiorina et al. (2003, 2010); Auzillon et al. (2011, 2012)), which are also commonly used in the combustion community, will not be described in the following.

Multiple approaches for chemistry description

Assembling a complete set of elementary reactions for a global combustion process results in a detailed chemical kinetic mechanism. It is important to note that such chemical schemes are still considered as models (see S. (2003)) since that reaction rate expressions still rely heavily on intuition: 1) the Arrhenius formulation is an approximation; 2) an optimization procedure is applied in all/some rates constants to match targets from the literature; 3) a large sensitivity of the results on some rates can be observed. These mechanisms are generally validated against experimental data for a large range of operating conditions and can therefore accurately reproduce a number of combustion processes. They are in practice used in chemical codes, like CANTERA Goodwin et al. (2017), or some DNS codes Patnaik et al. (1989); Tanahashi et al. (2000); Arani et al. (2017) mainly for hydrogen combustion because of the low number of species and reactions involved. Recently DNS of a lean methane/air turbulent premixed flame has been performed (Aspden et al. (2016)) using a detailed chemistry description. However, the use of detailed chemical schemes is still too expensive to be considered in most realistic configurations for most hydrocarbon fuels. The problem is the high number of reactions and species involved which increases drastically the computational cost of the simulation. It is possible to considerably reduce the number of species and reactions in these chemical schemes by removing the species and the reactions that are irrelevant to the problem considered. The validity of the skeletal mechanism obtained is then restricted to a certain range of operating conditions. It is also possible to replace the transport equation of some species with negligible production rate by an algebraic expression for their mass fraction. Because it is much less expensive to compute an analytical expression instead of solving a transport equation, this procedure yields an important reduction of the global cost of such mechanisms. The latter are called ARC for Analytically Reduced Chemistries and are employed in this thesis to perform flame inhibition simulations. The reduction procedure will be detailed in Part III. In the CFD community, ARC schemes have been used in cases where considering radical species is either mandatory or can considerably improve the simulation results (see Jones and Prasad (2010); Franzelli et al. (2013); Schulz et al. (2017); Jaravel et al. (2017)).



Figure 2.1: Validity span of different approaches to kinetic modeling.

Until recently, a common approach to combustion modeling for CFD was the use of **Globally reduced chemistries** (GRC), usually composed of a number of species lower than 10 and a number of reactions lower than 4 (Jones and Prasad (2011); Franzelli et al. (2013)). The idea behind these mechanisms is to reproduce global parameters (flame speed s_L or induction times during ignition for example) that are assumed to dominantly control the physics during the simulation. They are relatively simple, can be tuned easily to match certain targets and are cheap.

Fig. 2.1 displays a sketch of the validity span for the different approaches to chemical modeling discussed in this section.

2.5 Computational approaches for reactive Navier-Stokes equations

Finding analytical solutions to the Navier-Stokes equations (2.7-2.10) is, in most cases, impossible. Discretizing the different operators on a computational grid remains the main, if not the only, way to deal with this set of equations in three-dimensional configurations. The main difficulty with the numerical approach is the grid resolution needed to accurately solve the Navier-Stokes equations. This resolution depends on the range of time and length scales of the problem considered. An important issue when dealing with turbulent reactive flows, like explosions for example, is that this range can be quite large and is dictated by the coupling of turbulence and combustion processes. Even without combustion, turbulence itself is one of the most complex phenomenon in fluid mechanics. Indeed, turbulence can be characterized by fluctuations f' of local properties f. The latter can then be split into mean \overline{f} and fluctuating contributions: $f = \overline{f} + f'$. The turbulence fluctuations are associated with different scales ranging from the largest one, the integral length scale l_t , to the smallest one, the Kolmogorov scale η_k . l_t is usually close to the characteristic size of the geometry and defines the largest structures of the flow dominantly controlled by inertia. For structures smaller than l_t , the viscous forces become more and more important until they balance inertia at the Kolmogorov scale η_k . Numerical simulations of turbulent reactive flows may be achieved using three levels of resolution:

• When the grid resolution is sufficient to capture all the length scales of the flow, no model is needed and the approach is called DNS for Direct Numerical Simulation. DNS is very costly due to the number of points needed to capture both flow and flame structures. Even though the field of High Performance Computing (HPC) has



Figure 2.2: Turbulent energy spectrum as a function of wave numbers. (a) RANS, LES and DNS are conceptualized in terms of spatial frequency range. k_{l_t} , k_c and k_{η} are the wave numbers associated with the integral, cut-off and Kolmogorov length scales respectively.

known considerable break-through, DNS is still limited to canonical problems. DNS is however very useful to gain insight into complex mechanisms and the interplay among them, especially when a detailed description of the flow is needed. DNS will be used in this PhD to investigate the intricate DDT and inhibitor-flame interaction processes.

- On the other hand, when the averaged Navier-Stokes balance equations are solved, the approach reduces to RANS, for Reynolds Averaged Navier Stokes. The RANS method computes the mean quantities of interest, which requires closure rules: a turbulence model for the flow dynamics and a turbulent combustion model to describe heat release and chemical species conversion. RANS techniques are historically the first considered since solving the instantaneous turbulent flow was impossible. They are still the standard approach in all commercial codes for combustion today because of their low computational cost.
- In between RANS and DNS, there is LES for Large Eddy Simulation. This method consists in filtering the instantaneous balance equations using a cut-off length scale l_c , associated with wave numbers k_c in Fig. 2.2. The large scales of turbulences, namely larger than l_c , are explicitly calculated whereas the effects of smaller ones, namely smaller than l_c , are modeled using subgrid closure rules. Because of its reasonable cost compared to DNS, LES has become the standard research tool in the turbulent reactive flows community and is more and more used in the industry (during the motor design process for example). Note that LES is designed to tend toward DNS when the cut-off length scale, l_c goes to zero. In this thesis, LES is used to perform flame acceleration simulations. LES equations and closure models are described in details in the next section.

2.6 Large Eddy Simulations (LES)

LES methods consider a filtering operation performed on the Navier-Stokes balance equations. Let the filter size be Δ . The filtering operation on a quantity f is given by:

$$\overline{f} = \int f(x')G_{\Delta}(x - x')dx'$$
(2.25)

where G_{Δ} is the filter kernel (low-pass filter). The contribution of the subscale structures, i.e. structures smaller than the filter size Δ , is then given by: $f' = f - \overline{f}$. For compressible flows, a mass-weighted Favre Filtering is preferred:

$$\overline{\rho}\widetilde{f} = \int \rho f(x')G_{\Delta}(x-x')dx' \qquad (2.26)$$

At this point it is important to note that: 1) independently of the filtering operation, the filtered value of LES fluctuations $\overline{f'}$ is not zero and filtering a filtered value $\overline{\overline{f}}$ does not equal \overline{f} ; 2) filtering the Navier-Stokes equations requires to commute integral and partial differential operators, which is not valid under most conditions. The error related to this exchange of operators is however neglected in general.

Filtered Navier-Stokes equations

Filtering the instantaneous balance equations leads to the following equations:

$$\frac{\partial \overline{\rho}}{\partial t} + \frac{\partial}{\partial x_i} (\overline{\rho} \widetilde{u_i}) = 0 \qquad (2.27)$$

$$\frac{\partial}{\partial t}(\overline{\rho}\widetilde{u_i}) + \frac{\partial}{\partial x_i}(\overline{\rho}\widetilde{u_i}\widetilde{u_j}) = \frac{\partial}{\partial x_i}\left(\overline{\tau_{i,j}} - \overline{\rho}(\widetilde{u_i}\widetilde{u_j} - \widetilde{u_i}\widetilde{u_j})\right) - \frac{\partial\overline{p}}{\partial x_i}$$
(2.28)

$$\frac{\partial}{\partial t}(\overline{\rho}\widetilde{E}) + \frac{\partial\overline{\rho}\widetilde{u_i}\widetilde{E}}{\partial x_i} = -\frac{\partial}{\partial x_i}\left(\overline{\rho}(\widetilde{u_i}\widetilde{E} - \widetilde{u_i}\widetilde{E})\right) + \frac{\partial}{\partial x_i}\left(\overline{\lambda}\frac{\partial\overline{T}}{\partial x_i}\right)\right) - \frac{\partial}{\partial x_i}\left(\overline{\lambda}\frac{\partial\overline{T}}{\partial x_i}\right) + \overline{\dot{Q}} + \overline{\dot{\omega}}_{\overline{T}}$$
(2.29)

$$-\frac{\partial}{\partial x_i} \left(\sum_k h_{s,k} J_{i,k} \right) - \frac{\partial}{\partial x_i} \left(u_i (p \delta_{i,j} - \tau_{i,j}) \right) + \dot{Q} + \overline{\dot{\omega}_T}$$
(2.29)

$$\frac{\partial}{\partial t}(\overline{\rho}\widetilde{Y_k}) + \frac{\partial\overline{\rho}Y_k\widetilde{u_i}}{\partial x_i} = -\frac{\partial}{\partial x_i}\left(\overline{\rho}(\widetilde{u_i}Y_k - \widetilde{u_i}\widetilde{Y_k})\right) - \frac{\partial J_{i,k}}{\partial x_i} + \overline{\dot{\omega}_k}$$
(2.30)

Species and enthalpy laminar fluxes are generally negligible compared to turbulent fluxes. They can be either neglected or modeled using a simple gradient assumption:

$$\overline{J_{i,k}} = -\overline{\rho} \left(\overline{D_k} \frac{W_k}{\overline{W}} \frac{\partial \widetilde{X_k}}{\partial x_i} - \widetilde{Y_k} \widetilde{V_i^c} \right)$$
(2.31)

$$\overline{\lambda}\frac{\partial T}{\partial x_i} = \overline{\lambda}\frac{\partial \widetilde{T}}{\partial x_i} \tag{2.32}$$

$$\overline{\sum_{k} h_{s,k} J_{i,k}} = \sum_{k} \widetilde{h_{s,k}} \overline{J_{i,k}}$$
(2.33)

where $\widetilde{V_i^c} = \sum_k^N \left(\overline{D_k} \frac{W_k}{\overline{W}} \frac{\partial \widetilde{X_k}}{\partial x_i} \right)$. In addition, transport properties are also estimated using resolved quantities: $\widetilde{\mu} = \mu(\widetilde{T}), \ \overline{D_k} = \overline{\mu}/(\overline{\rho}Sc_k)$ and finally $\overline{\lambda} = \overline{\mu}\overline{C_p}(\widetilde{T})/Pr$. A number of other terms in equations (2.27-2.30) can not be computed directly in LES and require closure models. This is discussed in the following.

Reynolds stress tensor $\mathcal{T}_{i,j} = \widetilde{u_i u_j} - \widetilde{u_i u_j}$

This term is closed using a turbulence model. According to the Bousinesq assumption, unresolved momentum fluxes can be expressed as (Pope (2001)):

$$\mathcal{T}_{i,j} = -\mu_t \left(\frac{\partial \widetilde{u_i}}{\partial x_j} + \frac{\partial \widetilde{u_j}}{\partial x_i} - \frac{2}{3} \delta_{i,j} \frac{\partial \widetilde{u_k}}{\partial x_k} \right)$$
(2.34)

where μ_t is the subgrid scale viscosity. The different models for $\mathcal{T}_{i,j}$ differ in the expression for μ_t . In this thesis, LES simulations were performed using the WALE model Nicoud and Ducros (1999). This model has a number of advantages:

- a correct asymptotic behavior at the walls;
- no subgrid viscosity is applied in the case of pure shear.

The kinematic turbulent viscosity in the WALE model is given by:

$$\nu_t = (C_w \Delta)^2 \frac{(s_{i,j}^d s_{i,j}^d)^{3/2}}{(\widetilde{S_{i,j}})^{5/2} + (s_{i,j}^d s_{i,j}^d)^{5/4}}$$
(2.35)

$$s_{i,j} = \frac{1}{2} (\widetilde{g_{i,j}}^2 + \widetilde{g_{j,i}}^2) - \frac{1}{3} \widetilde{g_{k,k}}^2 \delta_{i,j}$$
(2.36)

where $C_w = 0.4929$ is a constant of the model, $\widetilde{g_{j,i}}$ is the resolved velocity gradient tensor and $\widetilde{S_{i,j}}$ is the symmetric part of the velocity gradient tensor.

Species and enthalpy turbulent fluxes

These therms are usually evaluated using resolved gradients:

$$\overline{\rho}(\widetilde{u_iY_k} - \widetilde{u_i}\widetilde{Y_k}) = -\overline{\rho}\left(\overline{D_k^t}\frac{W_k}{\overline{W}}\frac{\partial\widetilde{X_k}}{\partial x_i} - \widetilde{Y_k}\widetilde{V_i^{c,t}}\right)$$
(2.37)

where the species turbulent mass diffusion is given by $D_k^t = \mu^t / Sc_k^t$, Sc_k^t being the species turbulent Schmidt number, and $\widetilde{V_i^{c,t}} = \sum_k D_k^t \frac{W_k}{W} \frac{\partial \widetilde{X_k}}{\partial x_i}$. In a similar manner, the following model is used for the turbulent enthalpy flux:

$$\overline{\rho}(\widetilde{u_i E} - \widetilde{u_i}\widetilde{E}) = -\lambda^t \frac{\partial T}{\partial x_i} + \sum_k \widetilde{h_{s,k}} \overline{J_{i,k}}^t$$
(2.38)

where the turbulent heat diffusion coefficient is given by $\lambda^t = \nu_t \overline{Cp}/Pr^t$, Pr^t being the turbulent Prandtl number.

Species chemical source term $\overline{\dot{\omega}_k}$:

This term is crucial in reactive turbulent flows computations. Closure models for $\overline{\dot{\omega}_k}$ are discussed in Section 3.3.3.
2.7 Numerical aspects

The AVBP solver

AVBP Gourdain et al. (2009) is a DNS/LES massively-parallel solver for the fully compressible reacting Navier-Stokes equations. The equations are solved explicitly on unstructured and hybrid grids. It relies on a cell-vertex discretization method and generally treats boundaries according to the Navier Stokes Characteristic Boundary Conditions (NSCBC) formalism Poinsot and Lele (1992).

Numerical schemes

Different numerical schemes can be used to solve equations 2.27-2.30. Their dissipation and dispersion properties are of paramount importance to accurately reproduce flow and flame structures. The convective numerical schemes used during this thesis are:

- The Lax-Wendroff (LW) scheme (Lax and Wendroff (1960)) is a finite volume scheme with an explicit single step time integration. It is second order accurate in time and space. It has a low computational cost.
- The Two-step Taylor Galerkin (TTGC) scheme (Colin and Rudgyard (2000)) is a finite element scheme, with an explicit two-step integration in time. It is third order accurate in space and time. TTGC has much better dispersion and dissipation properties than LW, which makes it well-suited for LES applications. It is however approximately 2.5 times more expensive than the LW scheme.

The diffusion operator is descretized using the finite element diffusion scheme 2Δ developed by Colin (2000).

Artificial viscosity

The fact that AVBP uses centered numerical schemes leads to simulations that are prone to point-to-point oscillations (wiggles) close to regions of steep gradients. To attenuate these spurious oscillations, two artificial viscosities are employed in practice:

- A background dissipation term $(4^{th} \text{ order hyperviscosity})$ attenuates the amplitude of the wiggles.
- A 2nd order viscosity term introduces artificial dissipation which smooths local gradients. A sensor, detecting strong deviations of variables from linear behavior, is employed to apply the 2nd order viscosity term only in regions where the sensor is triggered avoiding global dissipation of the solution. The sensor used in this thesis is the one introduced in Colin (2000).

Handling shocks

To properly deal with the strong shocks involved in DDT, the Cook and Cabot (2004) sensor is used to add a spectral-like viscosity to stabilize numerical solutions and reduce oscillations near discontinuities. The Cook and Cabot (2004) method consists in introducing an additional bulk viscosity in the viscous stress tensor (Eq. (2.11)):

$$\tau_{i,j}^{Cook} = \tau_{i,j} + k' \frac{\partial u_k}{\partial x_k} \delta_{i,j}$$
(2.39)

$$= (k' - \frac{2}{3}\mu)\frac{\partial u_k}{\partial x_k}\delta_{i,j} + \mu\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right)$$
(2.40)

A spectral-like and grid dependent model for k' is chosen:

$$k' = C^r \rho \Delta_x^{r+2} \widetilde{|\nabla^r \mathbf{S}|}$$
(2.41)

where Δ_x is the grid spacing and **S** is the strain rate tensor $\mathbf{S} = ((\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i})/2)_{i,j=1,3}$. $\nabla^r S$ is the polyharmonic operator denoting a series of Laplacians. For example, it corresponds to the biharmonic operator $\nabla^4 S = \nabla^2 (\nabla^2 S)$ for r = 4. The \tilde{f} operator denotes a truncated Gaussian filter similar to the one introduced in Eq. 2.25. For unstructured grids, the only feasible option is to consider a single Laplacian, so that, in AVBP, r is set to 2 with the model constant $C^2 = 5$.

Chapter 3

Theoretical concepts of premixed flames

3.1 Introduction

The goal of this chapter is to provide some theoretical aspects of premixed flames. First, intrinsic properties of premixed flames are introduced. We then focus on the dynamics of premixed flame fronts in the laminar and turbulent regimes of propagation. Finally, several approaches to turbulent combustion modeling are discussed.

3.2 Laminar premixed flames

3.2.1 Unstretched premixed flames

The unstretched premixed flame is a canonical case to understand fundamental combustion processes. It corresponds to the situation where fuel and oxidizer are mixed prior to combustion. Temperature, pressure and composition of the mixture define the initial conditions of such flames. To define the composition, the equivalence ratio is used:

$$\phi = s \frac{Y_F}{Y_O} = \left(\frac{Y_F}{Y_O}\right) / \left(\frac{Y_F}{Y_O}\right)_s$$

where Y_F and Y_O are the fuel and oxidizer mass fractions in the mixture. s is the mass stoichiometric ratio which reads:

$$s = \left(\frac{Y_O}{Y_F}\right)_{st} = \frac{\nu'_O W_O}{\nu'_F W_F}$$

where ν'_F and ν'_O are the fuel and oxidizer stoichiometric molar coefficients of the global reaction.

Flame structure

The first attempt to describe the flame structure is attributed to Zeldovich and Frank-Kamenetskii (1938) and remains the basis of most asymptotic methods. Considering an axis positioned in the reference frame of a one-dimensional flame results in a steady state

version of the Navier-Stokes balance equations. A number of assumptions can be used to further simplify the equations:

- combustion proceeds through a single irreversible exothermic reaction controlled by an Arrhenius law with a large activation energy;
- one of the reactive species is in excess, so that its concentration varies little and its consumption does not affect the reaction rate. Only one species can therefore be considered. The reaction can then be described by the relation $R \to P + Q$, in which the reactant R is transformed into products P with the release of chemical energy Q;
- species R and P have the same molecular weight W, constant heat capacity C_p , molecular diffusion coefficient D and Lewis number Le = 1.

These assumptions yield the following set of equations:

$$\frac{\partial \rho u}{\partial x} = 0 \quad \text{or equivalently} \quad \rho u = cst = \rho_u s_L \tag{3.1}$$

$$\rho_u C_p s_L \frac{\partial T}{\partial x} = \frac{\partial}{\partial x} \left(\lambda \frac{\partial T}{\partial x} \right) - Q \dot{\omega}_R \tag{3.2}$$

$$\rho_u s_L \frac{\partial Y_R}{\partial x} = \frac{\partial}{\partial x} \left(\rho D \frac{\partial Y_R}{\partial x} \right) + \dot{\omega}_R \tag{3.3}$$

The above assumptions may seem strong, but the resulting equations preserve the global features of flames: intense non-linear heat release, variable density and temperature. A schematic of the obtained flame structure is displayed in Fig. 3.1(left), where 3 regions can be distinguished:

- a preheat zone, where fresh gases are heated due to thermal fluxes;
- a reaction zone, where the rate of the reaction $R \rightarrow P + Q$ increases drastically. Reactant R is converted into products P leading to chemical energy release Q;
- a post-flame zone, where the temperature reaches the adiabatic equilibrium value.

These simple models $R \to P + Q$ provide a global description of the flame structure and are useful to study theoretically some global flame parameters.

Intrinsic flame properties

One-dimensional laminar premixed flames have a characteristic consumption speed, s_L (Eq. 3.1). The set of equations Eqs. 3.1-3.3 can be integrated asymptotically (i.e. for very large activation energies) to obtain an analytical expression for s_L Zeldovich and Frank-Kamenetskii (1938); Williams (1985); Ferziger and Echekki (1993). A simple scaling law for s_L can be deduced from an asymptotic analysis of the governing equations and shows that:

$$s_L \propto (D_{th}A)^{1/2}$$
 (3.4)



Figure 3.1: Premixed flame structure using: (left) the simplest chemical model $(R \to P + Q)$ which mimics reactants R consumption to form products P while producing chemical energy Q; (right) detailed chemistry where the whole set of bi-molecular reactions are considered and where intermediate species I_k are considered.

A first estimation of the flame thickness can be deduced from s_L via scaling laws:

$$\delta_{th} \propto D_{th}/s_L \propto (D_{th}/A)^{1/2} \tag{3.5}$$

 δ_{th} is called the diffusive thickness and can be evaluated without flame computation. There are other ways to define a flame thickness, based on the flame profile. The thermal thickness δ_L , for example, can be obtained from the temperature profile as:

$$\delta_L = \frac{T_b - T_u}{\max(|\frac{\partial T}{\partial x}|)}$$

where T_b and T_u are the burnt and fresh gas temperature respectively. T_b corresponds to the final temperature of the gas after all chemical energy has been released. At constant pressure, the following expression for T_b is obtained:

$$T_b = T_u + QY_F^u / C_p$$

where Y_F^u is the fuel mass fraction in the fresh gases.

Flame structure using detailed chemistry

Some phenomena can not be described by the thermal runaway of the simple model presented in the last section. Important notions, like ignition, require to take into account the detailed reaction scheme. Indeed, in real flames, reactants are not directly converted into products. Instead, intermediate species I_k (see Fig. 3.1(right)), which are absent from the initial composition of the mixture, are produced via usually endothermic initiation reactions and interact with the reactants via chain branching reactions. The intermediate species are finally transformed into stable products P_k by strongly exothermic chain-breaking reactions.



Figure 3.2: Flame speed definitions from Poinsot and Veynante (2005).

3.2.2 Physical properties of stretched premixed flames

Multiple definitions of the flame speed

The laminar flame speed s_L was introduced in the previous section when describing the one-dimensional flame in its own reference frame. s_L is therefore the speed at which the flame front is moving with respect to the fresh gases. It is possible to define other flame speeds, based on kinematic flame properties, following Fig. 3.2:

- The flame absolute speed $s_a = \mathbf{w} \cdot \mathbf{n}$ is the normal component of the flame front speed $\mathbf{w} = (w_i)_{i=1,3}$ in the absolute reference frame.
- The flame displacement speed $s_d = s_a \mathbf{w} \cdot \mathbf{n}$ is the flame front speed relative to the local flow velocity.

It is also possible to define a flame speed based on the rate of consumption of reactants in the direction normal to the flame front:

$$s_c = \frac{1}{\rho(Y_F^u - Y_F^b)} \int_{-\infty}^{\infty} \dot{\omega}_F dn \tag{3.6}$$

where Y_F^b is the fuel mass fraction in the burnt gases. s_c is therefore a global quantity along the flame front normal, contrary to s_d which can vary from fresh to burned gases due to flow dilatation. One can introduce the density weighted displacement speed $s_d^{\star} = \frac{\rho}{\rho_u} s_d$ to cancel out the effect of the variation of density through the flame.

For a laminar steady unstretched premixed flame, the following relations hold in the reference frame of the flame:

$$s_a = 0 \tag{3.7}$$

$$s_L = s_c = s_d^\star \tag{3.8}$$

Effect of stretch on the flame speed

Before tackling multi-dimensional laminar and turbulent flames, it is important to introduce the notion of stretch. Total flame stretch κ is defined as the temporal variation of a flame surface element (Matalon and Matkowsky (1982); Candel and Poinsot (1990)):

$$\kappa = \frac{1}{A} \frac{dA}{dt} \tag{3.9}$$

Following Chung and Law (1984), κ can be decomposed as:

$$\kappa = \nabla_t \cdot \mathbf{u} + s_d \nabla_t \cdot \mathbf{n} \quad \text{where } \mathbf{n} \text{ is the normal to the flame front}$$
(3.10)

where ∇_t denotes the tangential component of the ∇ operator. The first term corresponds to the stretch induced by gradients of the flow upstream of the flame. It is called strain rate and disappears if the flow is uniform. The second term corresponds to curvature induced stretch. It disappears if the front is planar.

The effect of stretch on the flame speed has been studied theoretically using the simple ZFK model (Zeldovich and Frank-Kamenetskii (1938)). It is shown that the flame displacement speed s_d and the flame consumption speed s_c deviate from the laminar flame speed s_L linearly with respect to stretch κ for low stretch levels (Clavin (1985)):

$$\frac{s_d - s_L}{s_L} = -\mathcal{M}^d \frac{\kappa \delta_{th}}{s_L} \tag{3.11}$$

$$\frac{s_c - s_L}{s_L} = -\mathcal{M}^c \frac{\kappa \delta_{th}}{s_L} \tag{3.12}$$

In the above expressions, $\kappa \delta_{th}/s_L$ is a reduced Karlovitz number, and the coefficient \mathcal{M} is called the Markstein number. Neglecting the temperature dependence of the flame transport properties and assuming that the fuel is the deficient reactant, Clavin and Joulin (1983) obtained the following expressions for \mathcal{M} :

$$\mathcal{M}^{d} = \frac{v}{v-1}\ln(v) + \frac{1}{2}\beta(Le_{F}-1)\frac{1}{v-1}\int_{0}^{v-1}\frac{\ln(1+x)}{x}dx$$
(3.13)

$$\mathcal{M}^{c} = \frac{1}{2}\beta(Le_{F} - 1)\frac{1}{v - 1}\int_{0}^{v - 1}\frac{\ln(1 + x)}{x}dx$$
(3.14)

where β is the non-dimensional activation energy of the irreversible reaction and $v = T_b/T_u$ is the gas expansion ratio. From Eqs. 3.13-3.14, it appears that the Lewis number of the deficient reactant (taken as the fuel here) controls the flame speed response to stretch:

- For fuels with $Le_F \approx 1$, species and temperature gradients increase in the same proportions with increasing stretch. The flame becomes thinner and its overall consumption is not affected.
- For fuels with $Le_F < 1$ ($Le_F > 1$ resp.), the Markstein number is negative (positive resp.) and the consumption and displacement speeds increase (decrease resp.) linearly with increasing stretch.

Equations 3.11-3.12 imply that the total flame stretch is the only geometrical scalar controlling the normal burning velocity of a flame. This result dates back to Karlovitz et al. (1953) and is, in practice, far from being true. Indeed, since stretch decomposes into two contributions, one should expect two values for \mathcal{M} :

$$\frac{s_d - s_L}{s_L} = -\frac{\delta_{th}}{s_L} \left(\mathcal{M}_1^d \nabla_t . u + \mathcal{M}_2^d s_d \nabla_t . n \right)$$



Figure 3.3: A premixed flame, propagating away from the closed end of a wall, behaves as a semi-permeable piston.

The result $\mathcal{M}_1^d = \mathcal{M}_2^d = \mathcal{M}^d$ leads to Eq. 3.11 and is the consequence of the simplicity of the ZFK model. Numerical simulations using detailed chemical kinetics for methane combustion (Bradley et al. (1996)) have shown that \mathcal{M}_1^d and \mathcal{M}_2^d are different. Recently, Clavin and Graña-Otero (2011) performed an analytic study of flames sustained by a two-step chain-branching chemistry and confirmed that the two Markstein numbers are different. Keep in mind that these linear behaviors, for one or two Markstein numbers, hold only for low levels of stretch (namely $\kappa \delta_{th}/s_L \ll 1$) and a deviation from the linear law can be expected when exiting the low stretch region (see Poinsot and Veynante (2005)).

Effect of stretch on the burnt gas temperature

In absence of stretch, the burnt gas temperature T_b is equal to the adiabatic equilibrium temperature T_{ad} . According to Clavin and Williams (1982), stretch can induce differences between T_b and T_{ad} , provided that $Le_F \neq 1$ following:

$$\frac{T_b - T_{ad}}{T_{ad}} = \left(\frac{1 - Le_F}{Le_F}\right) \frac{D}{s_L^2} \kappa \tag{3.15}$$

where D is a characteristic diffusivity.

3.2.3 Propagating premixed flames: the shock-flame complex

Consider a canonical problem of interest in the gas explosion context: a planar premixed flame propagating away from the closed end of a tube (see Fig. 3.3). The density of the gas increases considerably through the flame from fresh to burnt states ($\rho_u/\rho_b \ge 5$ in most flames). In a coordinate system attached to the flame front, the fresh mixture enters the flame with a normal velocity s_L . According to Eq. 3.1, $\rho_u s_L = \rho_b u_b$ and the burnt gases leave the flame with a velocity $u_b > s_L$. Now, in the laboratory frame, the closed end of the tube corresponds to a zero normal velocity boundary condition, so that the burnt gases are at rest and the flame now moves down the tube with speed u_b pushing the fresh gases at speed $u_b - s_L$. Therefore, a premixed flame propagating away from the closed end of a tube acts like a semi-permeable piston.

The so-called piston effect is of crucial importance in the explosion phenomena. When a piston is set into motion in a tube filled initially with a gas at rest, the fresh gases are not set into motion instantaneously. Instead, the transition occurs progressively through the propagation of compression waves as illustrated in Fig. 3.4. The acoustic waves ahead



Figure 3.4: Illustration of the formation of the shock-flame complex for three increasing times $t_1 < t_2 < t_3$, when the propagation speed *a* is a function of the propagating quantity *u*.

of the flame eventually steepen and form a shock after a finite time, termed t_b for wave "breaking" ¹. For this reason, even in the planar case, a flame propagating away from a closed end usually consists of a reaction front and a precursor compression/shock wave.

3.2.4 Laminar flame acceleration mechanisms

Laminar flame acceleration in semi-confined/confined chambers

Freely propagating planar flames are observed only in special cases (flame propagating downwards in a vertical tube Searby and Quinard (1990); Pelce and Clavin (1982); Clavin and Garcia (1983)). In the cases considered in the present thesis, the explosion is initialized with a point source ignition from the closed end of a channel. The emerging initial flame is then spherical (or hemi-spherical). The reactive mixture is initially at rest. As mentioned in the introduction, we are not interested in open space explosions, and the walls can induce a strong flame acceleration during the early stages of the explosion. This has been suggested and studied experimentally by Clanet and Searby (1996). This mechanism is illustrated in Fig. 3.6 and discussed in the following:

¹The linearized version of the Euler equations predicts that the propagation of acoustic waves in a uniform medium occurs without deformation and at constant speed a_0 . Taking into account the non-linear terms in the Euler equations (by considering a propagation speed function of the propagating quantity a(u))), one can prove that there exists a finite time t_B at which multi-valued solutions will appear (using the Riemann Invariants Clavin and Searby (2016)).



Figure 3.5: Snapshot of the flame front in a narrow tube prior to the finger-flame regime. The flame front motion near the channel axis (i.e. $r \to 0$) is similar to a planar flame propagating away from a closed end. Equation (a) specifies the propagation velocity of a planar flame with respect to the burnt matter. On the other hand, close to the end wall (i.e. $X \to 0$), the flame motion is similar to a planar flame advancing towards the closed side wall. Equation (b) specifies the propagation velocity of a planar flame with respect to the fresh mixture. $u_{X,b}$ and $u_{r,u}$ are the projection of the flow velocity on the X and r axis respectively. The indices b and u denote the burnt and unburned matter respectively.

a) Spherical propagation. A few microseconds after ignition, a spherical flame forms. Like in the planar case if stretch effects are neglected, the flame tip displacement speed is constant and equal to $u_b = (\rho_u / \rho_b) s_L = \Theta s_L$. The flow ahead of the flame is pushed by the permeable piston at speed U_b .

b) Finger-flame. A clear understanding of the acceleration mechanism, subsequent to ignition, in the case of narrow tubes is attributed to the early work of Clanet and Searby (1996) and the extensive publications of Bychkov and coworkers (see in particular Bychkov et al. (2007) and Valiev et al. (2013)). Consider an initially spherical flame propagating from the closed end of a tube/channel of radius/half-width R with ideal slip walls. The analytical work considers the evolution in time of two particular points on the flame front: 1) the flame tip defined by the most advanced point on the flame front; 2) the flame skirt defined by one of the points where the flame is in contact with the end wall. Both points are displayed in Fig. 3.5. The equations describing their evolution in time is also recalled. The flame front near the channel axis (i.e. $r \to 0$) and the closed end wall (i.e. $X \to 0$) can be considered as planar.

• For $r \to 0$, the system reduces to a planar flame propagating away from a closed end. The evolution of the flame tip position X_{tip} can then be described using Eq. (a) in Fig. 3.5, which specifies the propagation velocity of a planar flame with respect to the burnt mixture. $u_{X,b}$ is the projection of the flow velocity, in the burnt gases, on the X axis.



Figure 3.6: Illustration of the formation of the finger shaped flame.

• On the other hand, close to end wall (i.e. for $X \to 0$), the flame motion is similar to a planar flame advancing towards the closed side wall. The evolution of the flame skirt position r_{skirt} can then be described using Eq. (b) in Fig. 3.5 which specifies the propagation velocity of a planar flame with respect to the fresh mixture. $u_{r,u}$ is the projection of the flow velocity, in the fresh mixture, on the r axis.

The whole system is closed once an expression for $u_{X,b}$ and $u_{r,u}$ is provided. Neglecting compressible effects and assuming a potential flow near the end wall and the channel axis, Bychkov et al. (2007) give the following forms for Eqs. (a) and (b):

$$\frac{dr_{skirt}}{dt} = s_L + \mathcal{C}(R^2 - r_{skirt}^2)$$
(3.16)

$$\frac{dX_{tip}}{dt} = \Theta s_L + 2\mathcal{C}r_{skirt}X_{tip} \tag{3.17}$$

where $C = (\Theta - 1)s_L/R^2$. The right hand side in both Eqs. 3.16 and 3.17 reduces to Θs_L when $r_{skirt} \rightarrow 0$ and $X_{tip} \rightarrow 0$ respectively. This means that Eqs. 3.16 and 3.17 reproduce the spherical regime in the early stages. As the flame skirt moves towards the side wall, the right hand side term of Eq. 3.17 increases, which implies an acceleration of the flame tip. An integration of the above system leads to the following expression for the flame tip position:

$$\frac{dX_{tip}}{dt} = \Theta s_L + \frac{2\alpha s_L \tanh(\mathcal{D}t)}{R} X_{tip}$$
(3.18)

with the solution
$$X_{tip} = \frac{\Theta R}{2\alpha} \sinh(2\mathcal{D}t)$$
 (3.19)

where $\mathcal{D} = \alpha s_L/R$ and $\alpha = \sqrt{\Theta(\Theta - 1)}$. Equation 3.19 predicts a quasi-exponential increase of the flame speed during the finger flame phase accompanied with a drastic increase of the flame surface. This acceleration mechanism is therefore strong and is responsible for the emergence of fast flames without the help of obstacles nor turbulence. This phase is illustrated in Fig. 3.6(a-c).

c) The flame touches the wall : When all the combustible trapped in the near-wall region is consumed, the flame reaches the wall. The FA mechanism is no longer supported and the flame decelerates. Using Eqs. 3.16 and 3.17, Bychkov et al. (2007) evaluated the position of the flame tip X_{tip}^{wall} at which the flame touches the wall (i.e. $r_{skirt} = R$): $X_{tip}^{wall} = \Theta R$.

The numerical simulations performed in this thesis will be confronted to the above theoretical results in Section 7.4.1. At this point it is important to note that the finger-flame



Figure 3.7: Flame shape for very lean ($\phi \approx 0.4$) (left) and stoichiometric (right) hydrogen/air explosions in confined chambers, from Katzy et al. (2016).

acceleration described above is Reynolds-independent and attributed to the zero normal velocity condition at the walls. Another mechanism, conceptually close to the Shelkin mechanism, has also been identified by Bychkov et al. (2005); Akkerman et al. (2006), when wall friction is taken into account. It was however only shown for relatively low flame Reynolds numbers $Re_f = s_L R/\nu < 110$. Contrary to the finger-flame, the Shelkin mechanism weakens with increasing Reynolds number and is not limited in time. The Shelkin mechanism is most likely to describe flame accelerations in very long smooth narrow tubes which is not the case in the numerical simulations performed in this thesis. For this reason, the theory behind the wall friction induced acceleration is not recalled here. Note that, at any moment during this early phase, two distinct instabilities (discussed in details in the book Clavin and Searby (2016)) can arise and induce wrinkling of the flame. The first instability is hydrodynamic and is termed **Darrieus-Landau (DL)** instability. It occurs as soon as the flame is submitted to local perturbations, at wavelengths larger than the flame thickness, forming convex and adjacent concave front sections. The change in density of the gas traversing the flame is responsible for the DL instability. In addition, thermo-diffusive effects can either stabilize or increase the growth rate of the DL instability depending on the Lewis number of the deficient reactant in the combustible (Le_{dr}) . If $Le_{dr} < 1$, the flame speed increases (decreases) in the parts of the flame front convex (concave) relative to the fresh gases leading to an additional amplification of the reaction front wrinkling (see Section 3.2.2 and the introduction of the Markstein number \mathcal{M}). These two instabilities eventually result in the formation of cellular flames as illustrated in Fig. 3.7 for very lean hydrogen/air flames.

In channels without obstacles, other mechanisms can induce a much stronger flame acceleration than the one discussed above (for example the formation of tulip flames Clanet and Searby (1996)). In this thesis, we are interested in obstructed channels and the effect of obstacles on flame propagation is discussed in the following.

Obstacle induced laminar flame acceleration

The presence of obstacles is known to enhance flame acceleration (Oran and Gamezo (2007); Ciccarelli and Dorofeev (2008)) generally by promoting the creation of a turbulent flow ahead of the flame. This will be discussed in Section 3.3 dedicated to turbulent



Figure 3.8: Flame-obstacle interaction. The OH-PLIF images and the flame speed evolution are extracted from Boeck et al. (2017b).

premixed flames. In the first phase of flame acceleration however, the flame speed is generally too low to observe the formation of turbulent structures and FA is laminar in nature as illustrated in Fig. 3.8. The fresh mixture, pushed by the flame, interacts first with the obstacle. The result is a strong velocity gradient just prior to the obstacle as shown in frame 1. When the flame approaches the obstacle, its closest part to the axis is convected with much higher velocity than the one close to the wall and facing the obstacle. During its propagation in the confined part, the flame is stretched and the burning rate increases, which in turn increases the flow speed ahead of the flame. This feedback loop induces a strong FA. When the flame tip gets past the recirculation zone created downstream of the flame, this confinement induced FA is no longer supported and the flame slows down.

3.3 Turbulent premixed flames

Up to now, the flows considered were laminar, in the sense that fluid particles travel along well-ordered neighboring layers. When the flow characteristic velocity exceeds a certain value, depending on the initial and boundary conditions, the flow exhibits strong mixing of the particles following a collection of eddies. The latter motion therefore appears random and is termed "turbulent".

The emergence of turbulent flames is generally associated with the enhancement of the flame acceleration process. First, the mechanism responsible for the generation of turbulence in obstructed channels is recalled in the following.



Figure 3.9: (a) Vortex shedding generated behind obstacles in the fast flame regime. (b) As the flame accelerates, the flow around the obstacles transitions from laminar (t_1) to turbulent (t_3) .

3.3.1 Generation of turbulence by piston effect in obstructed channels

As established in Section 3.2.3, a propagating flame acts like a piston pushing the fresh gases ahead of it. For confined and obstructed channels, walls and obstacles induce a non-uniform distribution of the fresh gases velocity ahead of the flame as illustrated in Fig. 3.8. Obstacles represent a source of perturbation for the incoming flow, promoting turbulence as illustrated in Fig. 3.9. In the early stages of the explosion, the flame acceleration is moderate so that the flow around the obstacles is laminar. As soon as the flame reaches a fast regime of propagation, the flow far ahead of the flame then propagates in a turbulent flow, characterized by a collection of eddies with a large range of sizes. The different regimes of turbulent combustion are identified by comparing the flow velocity fluctuations to the spatio-temporal scales of turbulence and laminar flames. This is discussed in Section 3.3.2.

3.3.2 Turbulent flame regimes

Once the self-propagating flame creates a turbulent flow ahead of its front, it can interact with the generated vortices. The impact of all these eddies on the flame shape and internal structure depends on the ratio of time scales between these structures and the flame.

In Fig. 2.2, two turbulent length scales have been introduced. The integral length scale l_t and the Kolmogorov length scale η_k correspond to the largest and smallest structures of the flow, respectively. The time scales corresponding to these structures can be obtained by taking into account the flow velocity fluctuations at scale l_t and η_k . Therefore, the integral and Kolmogorov time scales can be defined by $\tau_t = l_t/u'(l_t)$ and $\tau_{\mathcal{K}} = \eta_k/u_{\eta_k}$ respectively.

The combustion time scale can be estimated using the laminar flame speed s_L and the laminar flame thickness δ_L , hence $\tau_c = \delta_L/s_L$. The Damköhler number compares the



Figure 3.10: Turbulent combustion regimes (Peters (2000)) classified according to their position in the (u'/s_L) - (l_t/δ_L) diagram, along with the corresponding flame structure (Poinsot and Veynante (2005); Law (2010)).

integral time scale τ_t to the laminar flame time scale τ_c :

$$Da = \frac{\tau_t}{\tau_c} = \frac{l_t}{\delta_L} \frac{s_L}{u'(l_t)}$$
(3.20)

When $Da \gg 1$, the turbulent structures can wrinkle the flame but they are too large to impact the internal structure of the flame. In this **flamelet regime**, the flame remains locally laminar.

The Karlovitz number compares the smallest turbulent length scale to the reactive scales through:

$$Ka = \frac{\tau_c}{\tau_{\mathcal{K}}} = \frac{\delta_L \, u_{\eta_k}}{\eta_k \, s_L} \tag{3.21}$$

The turbulent Reynolds number $Re_t = l_t u'(l_t)/\nu$ links Da and Ka via:

$$Re_t = Da^2 K a^2 \tag{3.22}$$

Finally, another version of the Karlovitz number can be considered, based on the thickness of the reaction zone δ_r . The latter is generally orders of magnitude lower that the laminar flame thickness. This leads to:

$$Ka_r = \frac{\delta_r}{\eta_k} \frac{u_{\eta_k}}{s_L} < Ka \tag{3.23}$$

The combustion regimes can be decomposed following the diagram of Fig. 3.10:

Laminar flames ($Re_t < 1$): The flow is quasi-laminar. Hydrodynamic and thermodiffusive instabilities can still wrinkle the flame (tulip flames and cellular flames for example) but the resulting structure of the front is not attributed to the interaction of the flame with a turbulent flow. In this sense, the flame is considered laminar. In this thesis, this regime is encountered in the first phases of flame propagation, i.e. generally before it reaches the first obstacle.

Turbulent flames $(Re_t > 1)$

- Thin flames (Ka < 1): The flame thickness is smaller than the smallest turbulent structures, which can not interact with the internal structure of the flame. Hence, the latter remains locally laminar. The interaction of the large structures of the flow with the reactive front depends on the turbulence intensity:
 - Wrinkled flamelets ($Ka < 1, (u'/s_L) < 1$): Because turbulent fluctuations are moderate, the flame surface is only slightly wrinkled and stretched by the flow large structures. These combustion fronts are observed, in this thesis, when the flame interacts with the first obstacles.
 - Corrugated flamelets ($Ka < 1, (u'/s_L) > 1$): The turbulent fluctuations are higher and the flame surface endures high wrinkling and stretch. This typically corresponds to what can be observed when the flame propagates in a fully turbulent flow.
- Reaction sheets $(Ka > 1, Ka_r < 1)$: In this case, the smallest eddies (of size η_k), are able to modify the properties of the flame preheat zone (mass and heat transfer). The reaction zone, however, keeps its laminar structure. This regime can coexist with the previous one, in the last stage of turbulent flame propagation.
- Well-stirred reactors $(Ka > 1, Ka_r > 1)$: The whole flame internal structure is modified. This regime is not of interest for the cases studied in this thesis.

It is important to note that this classification is only qualitative and suffers from a number of assumptions that are far from being valid in practice (the idealized homogeneous isotropic turbulence, no interaction between heat release and turbulence, steady state consideration, single-step irreversible reaction...).

3.3.3 Turbulent combustion modeling and closure models for the subgrid turbulence-flame interaction

The filtering operation introduced by the LES formalism leaves unclosed chemical source terms, which have not been discussed yet. Since they can be very sensitive to the unresolved scales of turbulent motions, their closure is of critical importance in LES and is still a very active field of research in the LES community.

In this thesis, both single-step and detailed chemical schemes have been used. Consequently, we are interested in the approaches able to handle complex chemistry effects. A description of different methods available in the literature is proposed in the following.

3.3.3.1 PDF methods

In the PDF method, the fundamental idea is that the fluctuations lost by the filtering procedure can be described by introducing a probability density function $\mathcal{P}(\Phi)$ in the

phase space Φ , which describes the chemical state of the system: $\Phi = (Y_1, ..., Y_N, T)$ (Pope (1994)). The filtered source term can be reconstructed following:

$$\tilde{\dot{\omega}} = \int \dot{\omega}(\Phi) \mathcal{P}(\Phi) d\Phi \tag{3.24}$$

The different approaches to the PDF methods depend on their answer to the question of how the PDF can be obtained:

- The first approach is to presume a PDF shape. Many authors have proposed PDF functions that are summarized in Borghi (1988); Bray et al. (1989). The most common one, the βPDF , is based on the β function adapted to LES by Cook and Riley (1994). While this method has been extensively used combined with tabulated chemistry, its applicability to Arrhenius based chemistries (like the chemical schemes discussed in this thesis) is doubtful. Indeed, detailed chemistries involve a number of species that are strongly correlated, making the PDF shape hard to assume and difficult to decompose into canonical PDF functions. Another limitation to this method is that the predicted flame speed does not reduce to the laminar flame speed in absence of turbulence, as pointed out by Fiorina et al. (2010). Such a method is therefore not suited for the cases discussed in this thesis since we are interested in the case where, after ignition, the flame emerges laminar. Hence the need to correctly reproduce the laminar flame propagation.
- In the second approach, called stochastic PDF, the PDF is no longer presumed. Instead, transport equations are solved for the coarse grained PDF arising from Boltzmann equations (Pope (1994)). Such method yields an excellent description of the subgrid scale interactions. The main limitation comes from the complexity of the implementation and the associated cost.

3.3.3.2 Conditional Moment Closure

The CMC approach (Klimenko and Bilger (1999); Navarro-Martinez et al. (2005)) appears as an intermediate between the presumed PDF method and the full PDF method. A coarse grained PDF is introduced, but this time the phase space is represented by the mixture fraction. It reads:

$$\mathcal{P}(\eta, x, t) = \int \delta\left(\eta - \zeta(x', t)\right) G(x' - x, \Delta) dx'$$

where ζ is the fine grained mixture fraction. Finally, the scalars describing the chemical system are conditioned on η as:

$$Y_k(\eta, x, t) = \frac{1}{\rho \mathcal{P}(\eta)} \int \rho Y_k \delta\left(\eta - \zeta(x', t)\right) G(x' - x, \Delta) dx'$$

The transport equations are then solved for the conditioned scalars, and the unconditioned scalars are recovered following:

$$\widetilde{Y}_k(x,t) = \int Y_k(\eta, x, t) \mathcal{P} d\eta$$

Assuming that the statistical fluctuations decorrelated from η are small, one can then use a first order closure for the conditional source terms:

$$\dot{\omega}_k(Y_k, T) | \eta \approx \dot{\omega}_k(\langle Y_k | \eta \rangle, \langle T | \eta \rangle)$$

The probability density function is obtained by a β -PDF reconstruction from mixture mean and subgrid variance. To conclude, two points have to be made:

- This conditioning is interesting for some applications. For example, the ignition of a mixture occurs at the location of highest reactivity which can be translated to the mixture fraction;
- The number of equations to close and resolve can induce a very high computational cost. For this reason, many authors are forced to reduce the number of equations by explicitly taking into account the problem invariance.

3.3.3.3 Artificially Thickened Flame model

The Thickened Flame model (TFLES)

The main idea behind the LES approach is to resolve the large turbulent structures, while modeling the contribution of the small ones. Consequently, the typical grid resolution employed does not allow to resolve the flame thickness. The Thickened Flame model (TF) circumvents this problem by artificially thickening the flame front so that a certain number of computational cells are present inside the flame front. Recalling the asymptotic analysis of Section 3.2.1, it can be shown that the laminar flame speed and thickness scale as:

$$s_l \propto \sqrt{\dot{\omega}D}$$
 (3.25)

$$\delta_l \propto \sqrt{D/\dot{\omega}} \tag{3.26}$$

Therefore, if the diffusivities are multiplied by a factor \mathcal{F} such that $D \to \mathcal{F}D$ and the source terms are divided by the same factor (i.e. $\dot{\omega} \to \dot{\omega}/\mathcal{F}$), the flame speed is conserved while the laminar flame thickness is increased: $\delta_l \to \mathcal{F}\delta_l$. The thickening factor \mathcal{F} will then depend on the grid resolution needed to correctly resolve the flame thickness.

The problem with this technique is that it alters the flame interaction with turbulence as pointed out by Angelberger et al. (1998); Colin et al. (2000). Both the Da and Ka are modified by a factor \mathcal{F} and models need to be introduced to retrieve the contribution of the lost part of the turbulent spectrum (i.e. for sizes lower than $\mathcal{F}\delta_l$). At this point it is important to clarify what is meant by contribution. Following the approach of Poinsot et al. (1991), it is assumed that flow structures having sizes lower than a flame cut-off length scale $\delta_l^c \approx \delta_l$ do not alter the flame, mimicking the flamelet regime. Therefore, the flame is assumed to keep its laminar internal structure, and the approach only concerns the lost contribution to front wrinkling. The reduction of the flame surface leads to the decrease of the turbulent flame speed defined by: $s_T = (A_T/A)s_L$, where A_T/A is the ratio of the total wrinkled flame surface to the average surface (i.e. without the contribution of wrinkling). To compensate for this effect, an efficiency function \mathcal{E} is introduced. It is important to define which interaction is designated here to avoid any confusion. \mathcal{E} is defined as the ratio between the wrinkling of the unthickened flame and the wrinkling of the thickened flame:

$$\mathcal{E} = \frac{\Xi(\delta_L)}{\Xi(\mathcal{F}\delta_L)} \tag{3.27}$$

Multiplying both the diffusivities and the source term by \mathcal{E} leads to the increase of the flame speed $s_L \to \mathcal{E}s_L$, without impacting the flame thickness.

It leaves the question of how to describe \mathcal{E} . Various expressions have been proposed in the literature (Colin et al. (2000); Charlette et al. (2002a,b); Wang et al. (2011, 2012)). In this work, only the Colin et al. (2000) expression for Ξ is used and recalled here. It relies on the assumption of equilibrium between turbulence and subgrid scale flame surface. Ξ depends on the filter size Δ and is expressed as follows:

$$\Xi_{\Delta} = 1 + \alpha \frac{\Delta}{s_L} < a_T^{sgs} > \tag{3.28}$$

where $\langle a_T^{sgs} \rangle$ is the subgrid scale strain rate, i.e. the contribution of the structures smaller than the filter size Δ_e to the strain rate. $\langle a_T^{sgs} \rangle$ can be retrieved from the resolved turbulent velocity fluctuations at the filter scale u'_{Δ_e} as follows:

$$\langle a_T^{sgs} \rangle = \Gamma\left(\frac{\Delta}{\delta_L}, \frac{u'_{\Delta}}{s_L}\right) \frac{u'_{\Delta}}{\Delta}$$
 (3.29)

where Γ is the integral of the strain rate on scales ranging from the flame cut-off length scale to the filter size. To recover the Damkhöler theory (i.e. $\Xi \approx 1 + u'/s_L$) when the flame is wrinkled by all turbulent scales, α is estimated by:

$$\alpha = \beta \frac{\ln(2)}{2c_{ms}(Re_t^{0.5} - 1)}$$
(3.30)

where $c_{ms} \approx 0.28$ is a constant, $Re_t = u'l_t/\nu$ is the turbulent Reynolds number and β is the constant of the model. Finally, one obtains \mathcal{E} :

$$\mathcal{E} = \frac{\Xi_{\Delta_e}(\frac{\Delta}{\delta_L}, \frac{u'_{\Delta}}{s_L}, Re_t)}{\Xi_{\Delta}(\frac{\Delta}{\mathcal{F}\delta_L}, \frac{u'_{\Delta}}{s_L}, Re_t)}$$
(3.31)

Estimation of u'_{Δ_e} The estimation of the turbulent velocity fluctuations at the filter scale Δ_e is based on the resolved velocity u according to Colin et al. (2000):

$$u'_{\Delta_e} = |2\Delta_x^3 \nabla^2 (\nabla \times u)| \tag{3.32}$$

It is worth noting that recent numerical investigations of flame acceleration in semiconfined and obstructed channels (Quillatre et al. (2013); Vermorel et al. (2017)) showed that both Charlette et al. (2002a) and Colin et al. (2000) efficiency models give a satisfactory agreement with the experimental results for various fuels and explosion configurations provided that an appropriate model constant is chosen. The Colin et al. (2000) model has been retained for the numerical investigation presented in Chapter 7.

Note that a dynamic wrinkling formulation also exists (Wang et al. (2011)). It was successfully applied on turbulent jet flames (Volpiani et al. (2016)), on an aeronautical burner geometry (Volpiani et al. (2017b)) and on semi-confined gas explosions (Volpiani et al. (2017a)). This technique consists in automatically adjusting the flame wrinkling factor from the knowledge of the resolved scales. The main advantage of this dynamic approach, compared to the Colin et al. (2000) formulation for example, is that it does not assume equilibrium between turbulence and flame wrinkling. It was found to be relatively insensitive to numerical input coefficients and the additional cost introduced by the dynamic procedure is acceptable (around 5 - 10% of the total computation cost). However, for chronological reasons (the computations being performed before the study of Volpiani et al. (2017a)), this model was not used in this thesis.

The Dynamic Thickened Flame model (DTFLES)

In the initial formulation of the TFLES model, the thickening factor \mathcal{F} was applied everywhere in the domain. The diffusion coefficients were then modified even in regions where no chemistry is involved. This can modify the diffusion properties of the mixture in regions of crucial importance like in mixing zones or near the walls. To improve this aspect of the model, a dynamic ² procedure is employed in the DTFLES approach Legier et al. (2000). A sensor \mathcal{S} is therefore used to trigger the TFLES correction only in the flame region. The local thickening applied is expressed as:

$$\mathcal{F} = 1 + (\mathcal{F}_{max} - 1)\mathcal{S}$$

 \mathcal{F}_{max} is the maximum thickening determined locally by comparing the grid resolution to the laminar flame thickness:

$$\mathcal{F}_{max} = N_c \frac{\Delta_x}{\delta_L}$$

The control parameter N_c is roughly the number of cells used to "resolve" the flame front. For global reduced mechanisms, $N_c \approx 5$ in most situations and the sensor S is generally based on the fuel reaction, by introducing an Arrhenius-like expression:

$$\Omega = Y_F^{n_F} Y_O^{n_O} \exp(-\Gamma E_a / RT)$$

which is evaluated locally and compared to the value Ω_0 obtained from a 1D premixed flame calculation. The final expression for the sensor reads:

$$\mathcal{S} = \tanh(\beta' \Omega / \Omega_0)$$

with $\beta' \approx 50$. The quantity Ω has the form of a reaction rate, and the parameter $\Gamma = 0.5$ allows to trigger the sensor at lower temperature than the actual reaction in the scheme. This is done to capture the low temperature region of the flame.

²The term "dynamic" may be not suited for the model presented here since usually, when the authors talk about dynamic thickening, they refer to the capacity of the model to adapt the efficiency coefficient β in time and space. The authors recognize that use of the term "dynamic" here may be deemed abusive.

Chapter 4

Theoretical concepts of detonations

4.1 Introduction

Unlike premixed flames whose propagation is controlled by thermal conduction, the propagation of detonation fronts is governed by shock dynamics. The goal of this chapter is to provide some theoretical basis on detonation fronts. First, the detonation structure is discussed. We then focus on the dynamics of detonation fronts and the influence of multidimensionality on their propagation.

4.2 One-dimensional structure of detonations

As discussed in the introduction of this manuscript, a detonation corresponds to the supersonic mode of propagation of a combustion wave relative to the fresh gases. For this reason, shock dynamics controls the propagation of the detonation.

A shock wave corresponds to an abrupt (discontinuity for large Mach numbers) increase in the temperature and pressure of the mixture in which it propagates. When this mixture is reactive, the increase in temperature can be sufficient to initiate exothermic reactions. Under certain conditions, the reaction zone can remain attached to the shock to form a detonation. The structure of the latter is sketched in Fig. 4.1 for a detonation front propagating at constant speed D. The state of the gas downstream of the shock is called Von-Neumann (VN) and is denoted by the index N. The reactions are later initiated after an induction time τ_N , corresponding to an induction length $l_N = u_N \tau_N$, where u_N is the gas velocity downstream of the leading shock. The burnt gas state is denoted by the index b in the following.

The description of the internal detonation structure proceeds in three steps: 1) thermodynamic conditions at the VN state are evaluated using the jump conditions across the leading shock in Section 4.2.1; 2) thermodynamic conditions in the burnt gases are obtained, in Section 4.2.2, by considering the detonation as a reactive shock, i.e. bypassing all the intermediate states between fresh and burnt gases; 3) finally, the intermediate states between the leading shock and burnt gases are described by taking into account the reaction rate in Section 4.2.3.



Figure 4.1: Structure of a detonation front propagating at constant speed D. The state downstream of the leading shock is called Von-Neumann and is denoted by the index N in the following. After an indiction time τ_N , exothermic reactions are initiated. Once the combustion products are formed, the burnt gas state, denoted by the index b, is reached.

4.2.1 The Von-Neumann state: Rankine-Hugoniot relations

The conservation of mass in the reference frame of the detonation reads:

$$\rho_u D = \rho_N u_N = m \tag{4.1}$$

where m is the mass flow rate. Imposing a homogeneous velocity u everywhere except inside the shock, the momentum equation gives:

$$\rho_u D^2 + p_u = \rho_N u_N^2 + p_N \tag{4.2}$$

The sequence of states satisfying both equations (4.1) and (4.2) forms a line in the p - v space, where $v = 1/\rho$. This line is called the Rayleigh line:

$$\frac{p_u - p_N}{(1/\rho_N) - (1/\rho_u)} = \frac{p_u - p_N}{v_N - v_u} = -m^2$$
(4.3)

 $-m^2$ is therefore the slope of the Rayleigh line. The latter gives a family of solutions and the energy equation must be used to obtain the state N. Since no chemical energy has been released through the leading shock, the energy equation reads:

$$h_u + D^2/2 = h_N + u_N^2/2 \tag{4.4}$$

where h is the enthalpy. Using Eq. (4.3), one can transform Eq. (4.4) into:

$$h_N - h_u = \frac{1}{2}(v_N + v_u)(p_N - p_u)$$
(4.5)

For a polytropic gas, namely an ideal gas with constant specific heats per unit mass C_p and C_v , Eq. (4.5) now reads:

$$\frac{\gamma}{\gamma-1}\left(\frac{p_N}{\rho_N} - \frac{p_u}{\rho_u}\right) = \frac{1}{2}(v_N + v_u)(p_N - p_u) \tag{4.6}$$

$$(\gamma+1)\left(\frac{p_N}{p_u}-1\right)\left(\frac{v_N}{v_u}-1\right) = -2\left(\frac{p_N}{p_u}-1\right) - 2\gamma\left(\frac{v_N}{v_u}-1\right)$$
(4.7)

Introducing the notations,

$$\mathcal{P} \equiv \frac{\gamma + 1}{2\gamma} \left(\frac{p_N}{p_u} - 1 \right) \quad \text{and} \quad \mathcal{V} \equiv \frac{\gamma + 1}{2} \left(\frac{v_N}{v_u} - 1 \right)$$
(4.8)

Eq. (4.3) and Eq. (4.7) reduce to the following linear and equilateral hyperbola respectively in the $\mathcal{P} - \mathcal{V}$ space:

$$\mathcal{P} = -\mathcal{M}_u^2 \mathcal{V} \quad \text{and} \quad (\mathcal{P}+1) (\mathcal{V}+1) = 1$$

$$(4.9)$$

where $\mathcal{M}_u = D/a_u$ is the Mach number of the detonation relative to the fresh gases (a_u is the speed of sound on the reactants side). These two relations describe the Von-Neumann state. Note that the equilateral hyperbola has two asymptotes $\mathcal{P} = -1$ and $\mathcal{V} = -1$.

4.2.2 The burnt gas state

In a manner similar to Section 4.2.1 for shock waves, one can obtain the jumping conditions across a detonation front by skipping all the intermediate states between the fresh and burnt gases. The only difference lies in the presence of an exothermic reaction zone and Eq. (4.6) becomes:

$$\frac{\gamma}{\gamma - 1} \left(\frac{p_b}{\rho_N} - \frac{p_b}{\rho_u} \right) - \frac{1}{2} (v_b + v_u) (p_b - p_u) = q_m \tag{4.10}$$

where $q_m = C_p(T_b - T_u) + (u_b^2 - D^2)/2$ corresponds to the chemical energy released, per unit mass, once all chemical energy is release. Introducing,

$$\mathcal{P} \equiv \frac{\gamma + 1}{2\gamma} \left(\frac{p_b}{p_u} - 1 \right) \quad , \quad \mathcal{V} \equiv \frac{\gamma + 1}{2} \left(\frac{v_b}{v_u} - 1 \right) \quad \text{and} \quad \mathcal{Q} \equiv \frac{\gamma + 1}{2} \left(\frac{q_m}{C_p T_u} \right) \tag{4.11}$$

one can obtain the following relations:

$$\mathcal{P} = -\mathcal{M}_u^2 \mathcal{V} \quad \text{and} \quad (\mathcal{P}+1) (\mathcal{V}+1) = 1 + \mathcal{Q}$$

$$(4.12)$$

Since Q > 0, the present equilateral hyperbola lies above the one defined by Eq. (4.9) in the p - v plane. However, both curves have the same asymptotes.

Figure 4.2 displays the Rayleigh line and both Hugoniot curves (the two equilateral hyperbolae obtained by Eqs. (4.9) and (4.12)). The Neumann state N (burnt gas state b resp.) is given by the intersection of the Rayleigh line with the non-reactive Hugoniot curve (the Hugoniot curve after heat release resp.). Therefore, N and b depend on the slope of the Rayleigh line, which is given by the detonation propagation speed D:

• For a given D, the Rayleigh line intersects the Hugoniot curve after heat release if the following relation is satisfied: $(\mathcal{M}_u^2 - 1)^2 - 4\mathcal{Q}\mathcal{M}_u^2 \ge 0$. Therefore, a detonation is possible only if the detonation speed D is greater or equal than a threshold value D_{CJ} called Chapman-Jouguet detonation speed (equivalent to $\mathcal{M}_u \ge \mathcal{M}_{u,CJ}$). In other words, there is no solution for a steady planar detonation propagating at a constant speed smaller than D_{CJ} .



Figure 4.2: Sketch of a detonation in the p-v plane. The initial state corresponds to (p_u, v_u) . b is the burnt gas state and N is the Von-Neumann state.

- The CJ state corresponds to the case where the Rayleigh line is tangent to the Hugoniot curve after heat release. The burnt gas state b_{CJ} is therefore unique.
- For $D > D_{CJ}$, the Rayleigh line intersects the reactive Hugoniot curve in two points b and b'. The regime b is called strong detonation, whereas state b' is called weak detonation.

It is important to note that many investigators have ruled out the weak detonation solution (see Lee (2008); Clavin and Searby (2016)). Depending on the assumed path followed by the weak detonation solution in the p - v plane in Fig. 4.2, different arguments are provided:

- One can assume that the solution is directly reached from the initial state u along the Rayleigh line to the equilibrium state b'. In that case, Clavin and Searby (2016) argues that the thickness of a supersonic wave attached to a flame holder at room temperature would be non-realistic.
- One can also propose that the solution first undergoes adiabatic compression to reach VN before decaying to b'. In this case, authors either used entropy arguments (violation of the second law of thermodynamics) or stated that the structure is unstable (the flow behind a weak detonation is shown to be supersonic, thus any disturbance is unable to escape the detonation front). The second argument have been countered by Zeldovich (1940) by pointing out that an instability requires that the disturbances grow in amplitude with time and that the presence of disturbances at the detonation tail does not necessarily imply instability.

von Neuman (1942) did provide an argument for the possibility of a weak detonation solution, by constructing a structure later referred to as pathological detonation. The existence of such a wave has been confirmed by Dionne et al. (2000) for $H_2 - Cl_2$ mixtures. Therefore, it seems that the weak detonation solution is not so easily discarded.



Figure 4.3: Profile of flow velocity in the laboratory frame u_{lab} (i.e. where the fresh gas is at rest) for steady planar detonations. An overdriven detonation can be obtained by a piston placed downstream of the detonation, oriented in the direction of detonation propagation and moving at a speed v_p higher than $v_{p,CJ}$. Adapted from Clavin and Searby (2016).

4.2.3 From fresh to burnt gases: the ZND structure

The objective now is to describe the internal structure of a detonation (i.e for $D \ge D_{CJ}$). It is useful to introduce the progress variable c of the exothermic reaction. c = 0 corresponds to the fresh gases and c = 1 to the burnt gases. Similarly to the previous sections, one can describe the sequence of states from fresh to burnt gases by the following equations:

$$\mathcal{P} = -\mathcal{M}_u^2 \mathcal{V} \quad \text{and} \quad (\mathcal{P}+1) \left(\mathcal{V}+1\right) = 1 + c\mathcal{Q}$$

$$(4.13)$$

Equation (4.13) is a generalization of Eq. (4.12). It describes hyperbolae lying between the non-reactive Hugoniot curve (c = 0) and the Hugoniot curve after heat release c = 1. The detonation structure is found by expressing the flow velocity and the thermodynamic variables in terms of the progress variable c(x).

One can show that the flow behind the leading shock, in the reference frame of the detonation, is generally subsonic and the velocity increases with increasing c (i.e. through the detonation front). For $D > D_{CJ}$, the flow will remain subsonic through the entire detonation front. However, for $D = D_{CJ}$, the flow becomes sonic when the burnt gas state b_{CJ} is reached.

4.3 Selection mechanism of CJ detonations

Experimental investigations where self-propagating detonation waves are formed, either by direct initiation or via DDT, show that the detonation first propagates at velocities larger that D_{CJ} before decaying, after a sufficiently long time, to a quasi-steady value. In most cases, the experimental steady detonation propagation speed is very close to the



Figure 4.4: Selection mechanism leading to the CJ regime of autonomous detonation. The green part of the curves represents a rarefaction wave that emerges as soon as the piston is stopped. Adapted from Clavin and Searby (2016).

theoretical value D_{CJ} . Therefore, there must be a mechanism that activates once a detonation is formed to relax the solution towards the CJ regime. The answer lies in the downstream flow. As pointed out in the previous section, the flow downstream of a strong detonation is subsonic. Thus, an expansion wave can penetrate the reaction zone and attenuate the detonation. This is not the case when $D = D_{CJ}$ since the flow is sonic at the CJ plane (burnt gases). For this reason, detonations propagating at speed $D > D_{CJ}$ are unstable and can not be self-propagating. The CJ condition $D = D_{CJ}$ is the only solution that results in autonomous detonations. Nevertheless, strong detonations are observed before the CJ regime is reached and their existence is explained in the following. Consider steady detonation waves with a uniform flow after the end of the reaction zone

(see Fig. 4.3). In the reference frame of the detonation, the velocity at the burnt gases u_b is given by mass conservation through the detonation front $\rho_b u_b = \rho_u D$, where ρ_b is obtained using Eq. (4.12). In the laboratory frame, the boundary condition downstream of the reaction zone then corresponds to a piston pushing the burnt gases at a speed $v_p = D - u_b > 0$. Therefore v_p is given by $\rho_b(D - v_p) = \rho_u D$. v_p is minimum for $D = D_{CJ}$ and its value for a CJ detonation is $v_{p,CJ} = (1 - \rho_u/\rho_{b,CJ})D_{CJ}$. Strong planar solutions can be created by a piston whose velocity v_p is greater than $v_{p,CJ}$. For this reason they are called overdriven detonations. Finally, since D_{CJ} is the lower bound for detonation speeds, no supersonic combustion wave propagating at constant speed with a uniform flow between the end of the reaction zone and the piston can be observed for $v_p < v_{p,CJ}$.

Now, the question is what happens to an overdriven detonation when the system is no longer supported by a piston? When the latter is stopped, i.e. $v_p = 0$, a rarefaction wave \mathcal{R} forms and leaves the piston as illustrated in Fig. 4.4. Its speed is sonic relative to the burnt gases, so that its velocity in the laboratory frame is equal to $D - u_b + a_b$. Since the flow is subsonic $(u_b < a_b)$ in the overdriven detonation case, \mathcal{R} is faster than the detonation and can therefore reach the latter. When the rarefaction wave meets the detonation, the velocity of the latter decreases until it reaches the limit D_{CJ} . At this point, the rarefaction wave is blocked by the sonic plane (the velocity of \mathcal{R} is now equal to the CJ detonation speed D_{CJ}) and the disturbance can no longer penetrate the detonation



Figure 4.5: Typical soot record of a detonation (top) and associated hand tracing of the cellular structure (bottom). From Moen et al. (1982).

and reach the reaction zone. The detonation then propagates autonomously at constant speed D_{CJ} . This selection mechanism is of crucial importance since it explains why, in most experiments, the velocity of the detonation is found to be remarkably close to D_{CJ} after a sufficiently long time after onset.

4.4 Instabilities of detonation fronts

Despite the relatively steady propagation speed of self-sustained detonations, experimental studies generally reveal an unstable behavior of the detonation front, which appears when looking at the markings left on soot-coated foils on the walls. In the following, some aspects of unstable detonation fronts are discussed.

It was not until the early 1960s that experiments were carried out by D. (1961); W. (1961); S. (1965) revealing the detailed structure of unstable detonation fronts. The experimental technique uses soot-coated foils on the walls that leave a trace of the trajectories of different shocks. The markings left by the different shocks exhibit diamond or cellular like structures. A typical soot record is displayed in Fig. 4.5. These cellular patterns can be more or less regular depending on the explosive mixture. Nowadays, a huge database can be found online (see for example Detonation Database) recording the average dimensions of the detonations cells for various conditions and mixtures. This database is continuously updated thanks to extensive experimental investigations of detonation fronts behavior. Since the first observations, many authors have contributed to the fundamental understanding of the detonation instabilities. The underlying mechanism is related to the one-dimensional instability of the ZND structure combined with the transverse running singularities on the leading shock front, as explained in Sections 4.4.1 and 4.4.2 respectively.



Figure 4.6: One-dimensional instability of the ZND structure. l_I represents the length of the induction zone.

4.4.1 One-dimensional instability

In a one-dimensional framework, the instability of the detonation takes the form of a longitudinal pulsation in the direction of propagation. The one-dimensional instability of the ZND structure is due to the presence of the induction zone downstream of the leading shock and its sensitivity to changes in the thermodynamic conditions at the leading shock. This instability is sketched in Fig. 4.6. A disturbance that strengthens the leading shock reduces the induction delay and causes the reaction region to move upstream. The heat release zone then pushes the gas ahead of it, like a permeable piston. An acoustic pulse is then formed, runs upstream and reinforces the leading shock. A loop of upstream and downstream moving disturbances is then created.

It is important to note that the above disturbances, transmitted back and forth between the leading shock and the reaction zone, are not of the same nature. The upstream running perturbation is driven by an acoustic wave and propagates at the speed of sound. On the other hand, the disturbances propagating downstream are driven by both acoustic and entropy waves at the sound and flow speeds respectively. For this reason, there is a phase shift causing the instability to be oscillatory. Since the flow behind the leading shock is subsonic, the entropy wave imposes the longest delay in the loop.

The main parameter controlling the instability is the activation energy E_a , which represents the temperature sensitivity of the reaction. The degree of overdrive $f = D/D_{CJ}$ also plays a role by changing the dominant mechanism in the loop. For large values of f, Clavin and He (1996) show that the quasi-isobaric downstream propagation of the entropy waves controls the instability. Close to the CJ regime, i.e. for f close to unity, the instability is driven by the upstream running acoustic wave as shown by Clavin and Williams (2002).



Figure 4.7: Time evolution of a cellular detonation showing the traces of triple points trajectories. The propagation of these triple points coupled with the longitudinal pulsation of the ZND structure is responsible for the diamond pattern characteristic of detonations. From Clavin and Searby (2016).

4.4.2 Multidimensional diamond pattern

The cellular pattern observed in experiments (see Fig. 4.5) is due to a combination of the one-dimensional pulsating ZND structure and the formation of singularities on the leading shock front. These singularities are called triple points and are systematically observed on weakly perturbed shock fronts either by undulated walls (see Briscoe and Kovitz (1968)) or by interaction with vortices and turbulence (see Larsson et al. (2013)). These triple points propagate in the direction transverse to the shock at the speed of sound, strengthening the leading shock. They are visible at the location of the cusps illustrated in Fig. 4.7. These unstable modes depend on the mixture and the thermodynamic conditions. However, they do not appear instantaneously. Indeed, when the leading shock is perturbed, singularities form and the detonation can go through a number of unstable modes before reaching the steady mode observed in experiments of regular detonations. This selection mechanism was studied by Clavin and Denet (2002).

It is important to note that the relative importance of the longitudinal and transverse instabilities depends on the stable nature of the detonation and on the degree of overdrive (Clavin and Searby (2016)): a detonation can be stable with respect to longitudinal perturbations (i.e. stable ZND structure) and still be unstable with respect to transverse perturbations, hence leaving cellular patterns at its tail.

Chapter 5

Computation strategies for gas explosion simulations

All the simulations presented in this manuscript have been performed with the AVBP code. However, depending on the problem considered, different numerical strategies have been used. For the sake of clarity, we summarize here the specific methods selected for each case: FA, DDT and inhibited FA. They are also illustrated in Fig. 5.1.

5.1 Flame acceleration

The first part of the thesis investigates the mechanisms responsible for flame acceleration. The LES approach has been used to successfully compute small to large scale deflagrations with various fuels in the recent years (Ibrahim et al. (2009); Gubba et al. (2011, 2008, 2009); Zbikowski et al. (2008); Molkov et al. (2007); Wen et al. (2012); Xu et al. (2015); Zhao et al. (2017); Tolias et al. (2017); Volpiani et al. (2017a); Vermorel et al. (2017)). The LES method is employed in this work to compute FA in semi-confined and confined configurations.

Flame front propagation is dominantly controlled by the fuel burning velocity s_L , the gas expansion factor T_b/T_u and curvature effects controlled by the Lewis number of the deficient reactant Le_{dr} . GRCs are able to correctly reproduce these targets while being very simple as shown by Quillatre et al. (2013); Vermorel et al. (2017) for fuels like hydrogen, methane and propane. Therefore, a single-step mechanism is used in the first part of the thesis to compute FA.

5.2 DDT

DDT is triggered by a number of different scenarios occurring at small time and length scales. Understanding the different mechanisms involved requires to capture both flow and flame structures with high accuracy. Therefore, the DNS approach seems mandatory. Usually, (see the work of Oran and collaborators summarized in Oran and Gamezo (2007)) Adaptive Mesh Refinement (AMR) is used to add grid points in regions where accuracy is needed (flame front, detonation front, etc...) while decreasing the resolution in regions where the physics is controlled by large scale structures. This way AMR can drastically reduce the cost of the simulation. However, this method is mostly used in DNS codes



Figure 5.1: Overview of the different computational strategies used to simulate FA, DDT and Inhibited FA. DPS stands for Discrete Particle Simulation. We distinguish in this work FA, which is intrinsic to flame front propagation, from Inhibited FA where an external mitigative measure is applied on the flame path. The mechanisms observed during FA are also observed during Inhibited FA.

relying on cartesian grids. For unstructured grids, this kind of approach is much more difficult to implement. At the moment, such a method is not available in AVBP. For this reason, a simplified approach is adopted in this thesis. The mesh resolution is almost homogeneous and fine enough to capture both flame and flow structures. This method is therefore more costly than AMR but allows a detailed description of the onset of detonation in order to understand the intricate mechanisms involved. However, a three-dimensional DNS of the configuration considered is not feasible, thus only two dimensional simulations are performed.

The majority of the DDT simulations in the literature have been performed using a singlestep mechanism. According to Oran and Gamezo (2007), these mechanisms are designed to reproduce key features of flame and detonation propagation, while the transition from one regime to another (i.e. DDT) is not directly considered during the fitting procedure. Because detailed mechanisms are validated against experimental data for a large range of experimental targets and operating conditions, they can improve our understanding of DDT. In addition, they are affordable for fuels like hydrogen. For this reason, simulations of transition to detonation are performed in this thesis using both multi-step and singlestep kinetic schemes to assess the influence of chemistry modeling on DDT.

5.3 Inhibited FA

Similarly to FA, simulations of inhibited deflagrations are performed using LES for the gaseous phase. To take into account the solid phase, i.e. the inhibitor particles, a La-

grangian approach, also called Discrete Particle Simulation (DPS), is used to track the inhibitor particles. This Lagrangian formalism is detailed in Section 11.1. As regards chemical schemes, the complex inhibitor interaction with the flame chemistry requires to take into consideration the flame radical species depleted by the volatilization of the inhibitor particles. For this reason GRCs can not be used. However, the detailed description of methane combustion (fuel considered in this work) involves a high number of species (53 for the reference mechanism GRI3.0; Smith et al. (2017)) which makes it too costly to be considered for three-dimensional deflagration simulations. A reduction procedure (detailed in Section 11.2) is thus used to develop an ARC with a computational cost deemed affordable.

Part II

What are the mechanisms responsible for the extreme damages observed during VCE's?

Part II: What are the mechanisms responsible for the extreme damages observed during VCE's?

In this part of the manuscript, we assume that no mitigation procedure is available on the industrial plant and that the explosion starts with a mild ignition, i.e. a subsonic combustion wave emerges from the ignition source. We focus on the behavior of selfpropagating flames in confined and obstructed channels. The typical VCE scenario ends with the production of either fast flames or detonations. Because both FA and DDT are boundary dependent problems, a comparative study between these two scenarios is only meaningful if the same configuration is used. Chapter 6 presents two hydrogen/air explosion cases selected from the Gravent database (Vollmer et al. (2012)): first, a very lean mixture ($\phi = 0.52$) where only flame acceleration was observed and then a detonating stoichiometric case. The following investigation proceeds in three steps. First, FA alone is considered, in Chapter 7, to highlight the role of congestion and confinement in producing strong FA mechanisms and high overpressure levels. Then, Chapter 8 tries to explain why DDT was not observed in the case $\phi = 0.52$. An overview of different mechanisms that can explain DDT in the case $\phi = 1$ is also presented, which prepares the ground for the numerical study of the detonating case. Finally, Chapter 9 focuses on the key events surrounding the onset of detonation. It also shows how the DDT scenario can be influenced by the chemistry modeling.
The Gravent explosion channel

The Gravent explosion channel (Vollmer et al. (2012)) is operated at the Institute of Thermodynamics, Technical University of Munich. It is characterized by a high aspect ratio (length-to-width/diameter ratio). The basic idea is to provide a high degree of confinement and congestion to reach fast combustion regimes at laboratory scale. The channel is entirely closed and equipped with evenly spaced obstacles. The Gravent explosion channel offers a large database of gas explosion results available on the TUM website ¹. The impact of various parameters is investigated: obstacle shape, dimensions and spacing; combustible composition and transverse heterogeneities; water mist mitigation. In this thesis, we are only interested in the homogeneous cases without water mist. The study concerns only mixtures of hydrogen and air at various compositions.

The channels considered are composed of two distinct regions: a first zone equipped with a series of fence-type congestions followed by an obstacle free section that extends to the end plate. The design of such channels varies following 3 parameters:

- The degree of congestion at the obstacles characterized by the blockage ratio $\varsigma = 2h/H$, where h and H are the heights of an obstacle and the channel respectively;
- The spacing between obstacles S;
- The length of the obstructed region, i.e. the position of the last obstacle.

A schematic of the BR30hS300 channel, retained for this work, is displayed in Fig. 6.1. The dimensions of the channel are $L \times l \times H = 5400 \times 300 \times 60 \ mm^3$. 7 flat obstacles are placed on the bottom and top plates. The first obstacle is placed 250 mm away from the ignition plate. The spacing between obstructions is constant and equal to 300mm. The last obstacle is placed 2050 mm away from the ignition plate. What remains of the channel is obstacle free. All the obstacles have the same dimensions: $12 \times l \times h \ mm^3$, with h = 9mm. Therefore, the blockage ratio is: $\varsigma = 2h/H = 30\%$.

Note that the experimental apparatus, in the case of homogeneous mixture, is designed to study the effect of both confinement and obstruction on FA and DDT. When a detonation is triggered in the obstructed region, it later propagates in the large obstacle-free section where there is no geometrical perturbation. This allows to exploit the soot foils markings and to investigate the dynamics on such unstable fronts. However, when DDT is not observed, the flame propagation in the large obstacle-free section is not of significant relevance to FA.

¹http://www.td.mw.tum.de/en/research/ddt/



Figure 6.1: Schematic of the BR30hS300 configuration. The dimensions of the channel are $L \times l \times H = 5400 \times 300 \times 60 \ mm^3$. 7 flat obstacles are placed on the bottom and top plates. The first obstacle is placed 250 mm away from the ignition plate. The spacing between obstructions is constant and equal to S = 300 mm. The last obstacle is placed 2050 mm away from the ignition plate.

6.1 Experimental procedure

The channel is first filled with a hydrogen/air mixture at the desired composition. The combustible is then ignited at X = 0 using an electric spark.

The authors used various measurement techniques to analyze the experiment. We will rely mainly on the conventional measurements, i.e. time of arrival photodiodes and piezoelectric pressure transducers. These methods allow to detect the combustion front and the acoustic waves/shocks produced during the flame propagation in the channel. Therefore the experimental data include: 1) the time evolution of the flame tip (i.e. the most advanced point on the flame front) position inside the channel; 2) the pressure signal at 7 probes mounted at the top plate along the channel. The positions of the 7 pressure transducers are: $\{400, 1400, 2300, 3200, 4100, 5000, 5400\}$ mm. For each experimental setup, the results of several shots are provided. A very good reproducibility of the experimental results is observed.

6.2 Influence of the mixture composition on the explosion scenario

In the experiments (homogeneous cases), the volume fraction of hydrogen $X_{H_2}^{vol}$ ranges from 15% to 40%. For these mixtures, the laminar flame speed increases with increasing $X_{H_2}^{vol}$ as shown in Fig. 6.2(right). The speed s_{tip} of the flame tip at the end of the obstructed section, i.e. at X = 2050 mm, is displayed in Fig. 6.2(left). For $X_{H_2}^{vol}$ in [17.5 - 22.5]%, strong FA are observed resulting in flame propagation speeds much higher than 600 times the laminar flame speed s_L . In most cases this corresponds to supersonic flames² relative to the speed of sound of the reactants at initial conditions a_u^0 . For $X_{H_2}^{vol} \geq 25\%$, the experimental data suggests the formation of autonomous detonations in the obstructed

 $^{^{2}}$ Note that these combustion waves are still flames propagating at subsonic velocities relative to the reactants ahead of them. Their propagation velocity is supersonic relative to a fix point in the laboratory frame.



Figure 6.2: Left: Experimental flame tip velocity s_{tip} at X = 2050 mm, i.e. at the exit of the obstructed section, as a function of the hydrogen volume fraction in the combustible. Right: Laminar hydrogen/air flame speed s_L as a function of the hydrogen volume fraction. From Böck (2015). a_u^0 is the sound speed of the reactants at initial conditions.

section $(s_{tip} \text{ very close to } D_{CJ})$. Therefore, there exists a threshold value $[X_{H_2}^{vol}]_c$, in the range [22.5 - 25]%, above which DDT can occur.

For the numerical simulations presented in the next chapters two cases have been selected. The case $X_{H_2} = 17.5\%$ ($\phi = 0.52$) allows to investigate the mechanisms of FA involved in the BR30hS300 configuration. This case will be referred to as the "deflagrating case". DDT will be studied in the case $X_{H_2} = 30\%$ ($\phi = 1$) ("detonating case").

Chapter 7

LES of flame acceleration in a confined and obstructed channel

Abstract

The capacity of LES to correctly reproduce the key features of flame acceleration in complex configurations has been shown in numerous works (see Ibrahim et al. (2009); Gubba et al. (2011, 2008, 2009); Zbikowski et al. (2008); Molkov et al. (2007); Wen et al. (2012); Quillatre et al. (2013); Xu et al. (2015); Zhao et al. (2017); Tolias et al. (2017); Volpiani et al. (2017a); Vermorel et al. (2017)). These LES concern confined and semi-confined channels ranging from small to large scales filled with various fuels $(H_2, CH_4, C_2H_4, C_3H_8)$. Therefore, to perform the simulations of the deflagrating case, the LES approach is selected. First, the numerical strategy is summarized in Section 7.1. In Section 7.2, LES results are validated against experimental data in terms of flame speed and overpressure levels. The influence of the thermal boundary conditions on the flame propagation is investigated in Section 7.3 before a thorough analysis of the flame acceleration process is presented in Section 7.4. The BR30hS300 channel is complex, offering both confinement and congestion in the flame path. Therefore, there can be a number of mechanisms that can explain how an initially very subsonic combustion wave can reach a very fast propagation regime. The investigation reveals a combination of multiple FA mechanisms influencing the flame propagation. The chapter ends with a theoretical study of the flame journey through the congestion with the purpose of linking the flame acceleration rate through a sudden contraction to the flow contraction.

7.1 Numerical setup

7.1.1 Mesh

The BR30hS300 channel, presented in Fig. 6.1, is discretized using tetrahedral cells. The mesh is piecewise-uniform as shown in Fig. 7.1 The characteristic size Δ_x is equal to 2 mm in the regions free of obstacles (i.e. before the first obstacle, in the regions separating two successive obstacles and from the last obstacle to the end plate), which corresponds to 30 cells along the channel height. Around the obstacles, the resolution is increased ($\Delta_x = 1 \ mm$) to allow a good reproduction of the obstacle. The mesh is composed of



Figure 7.1: Schematic of the mesh used to discretized the BR30hS300 configuration. The mesh is composed of tetrahedral elements with a characteristic size $\Delta_x = 2 \ mm$ in the unobstructed areas. Around the obstacles, the resolution is increased ($\Delta_x = 1 \ mm$) to allow a good reproduction of the obstacle/flame interaction. The mesh is composed of approximately 70 million cells.

approximately 70 million cells. It is important to note that various meshing strategies, not detailed here, have been tested before this mesh was retained.

7.1.2 Boundary conditions

The channel is entirely closed so that all boundary conditions correspond to walls. A zero velocity condition is not suited for this simulation considering the mesh resolution employed $(y^+ \approx 150 - 200)$ and a law-of-the-wall (Schmidt et al. (2007)) is preferred. Both adiabatic and isothermal conditions have been applied on the walls to assess the influence of the heat fluxes on the explosion.

7.1.3 Initial conditions

The domain is filled with a quiescent very lean ($\phi = 0.52$) homogenous mixture of hydrogen and air. In the experiments, the explosion is initialized with an electric spark at the ignition plate X = 0. Modeling the ignition phase can be complex and is beyond the scope of the present study. Instead, the deflagration, in the LES, is initialized with an hemispherical flame:

- The radius of the flame kernel is 7mm. Abdel-Raheem et al. (2015); Volpiani et al. (2017a) have shown that for relatively low values (typically lower than 10mm), the initial flame radius does not impact the flame propagation characteristics.
- The internal structure of the initial flame is that of an artificially thickened laminar 1D flame at the operating conditions (the thickening is chosen so that 5 cells are present inside the thermal thickness).
- An initialization with a zero velocity field gave rise systematically to numerical noise, thereby strongly impacting the early stages of flame propagation. For this reason, a velocity profile based on the density profile through the flame is also imposed. Details on this procedure can be found in the thesis of Bonhomme (2014) (Section 1.6.2).

Reaction		Reaction rate model				A $[\mathrm{cm^3.mole^{-1}.s^{-1}}]$		$E_{a} \; [cal.mole^{-1}]$	
$H_2 + 0.5O_2 \rightarrow$	H_2O	$\mathbf{A}e^{-\mathbf{E}_{\mathbf{a}}/RT}[H_2]$			$[O_2]^{0.5}$	$8.59 10^{13}$		34966	
	H_2	O_2	H_2O	N_2			$\mathrm{s_L} \; [\mathrm{m.s^{-1}}]$	T_{ad} [K]	$\delta_{\mathbf{L}} [\mathbf{m}]$
Lewis numbers	0.3	1.11	0.81	1.13	Flame c	haracteristics	0.59	1657	$2.03 \ 10^{-4}$

Table 7.1: Single-step mechanism to model hydrogen/air chemistry at $\phi = 0.52$ and atmospheric conditions.

7.1.4 Chemistry

Several studies Ibrahim et al. (2009); Gubba et al. (2011, 2008, 2009); Zbikowski et al. (2008); Molkov et al. (2007); Wen et al. (2012); Quillatre et al. (2013); Xu et al. (2015); Zhao et al. (2017); Tolias et al. (2017); Volpiani et al. (2017a); Vermorel et al. (2017)) showed the capacity of single-step mechanisms to correctly predict the main features of flame acceleration as soon as key ingredients of flame propagation are reproduced. These chemistries are fitted against the laminar flame speed s_L and the adiabatic burnt gas temperature T_{ad} at the initial conditions of the explosion. These two targets allow to correctly reproduce fuel consumption and gas dilatation across the reactive front respectively.

For the LES of flame acceleration presented in this chapter a single-step approach is adopted. The chemical scheme involves 4 species $(H_2, O_2, H_2O \text{ and } N_2)$ and one irreversible reaction: $H_2 + 0.5O_2 \rightarrow H_2O$. An Arrhenius formulation is used for the reaction rate and takes the form: $\dot{\omega} = Ae^{-E_a/RT}[H_2][O_2]^{0.5}$, where the parameters A and E_a are given in Table 7.1. The Lewis number of the species are obtained from 1D premixed laminar flame computations at atmospheric conditions (at the reactants side) using the chemistry solver CANTERA Goodwin et al. (2017) and a detailed chemistry. This mechanism gives a laminar flame speed $s_L = 0.59 \ m/s$ within the range $0.54 - 0.63 \ m/s$ given by several detailed models for hydrogen/air chemistries. The adiabatic temperature $T_{ad} = 1657 \ K$ is also in good agreement with the one obtained with detailed mechanisms $T_{ad} = 1654 - 1659 \ K$. The laminar thermal flame thickness given by the model is around 200 μm .

7.1.5 Convection scheme

The numerical simulations have been performed using the Lax-Wendroff (LW) scheme Lax and Wendroff (1960). Attempts have been made to perform the simulations with more accurate schemes available in AVBP. However, a systematic blow up of the solution was observed when the flame reached high velocities (typically around $300 \ m/s$) between the third and fourth obstacles. The simulations could not be carried on until the end of the flame propagation in the obstructed region. Sudden accelerations of part of the flame gave rise to undershoots in the species profiles leading to the blow up of the solution. These acceleration induced undershoots are also observed when LW is used. However the latter is more dissipative than the other convection schemes, which allows to dump these oscillations in this case.

7.1.6 LES models

The characteristic size of the mesh used in the simulations of flame acceleration is around 2 mm, which is one order of magnitude higher than the laminar flame thickness δ_L . The DTFLES model, described in Section 3.3.3.3, circumvents this limitation by thickening the flame front. The value of the thickening factor \mathcal{F}_{max} in the flame front region depends on the number of cells N_c specified by the user to remain inside the thermal thickness of the thickened flame. In the following, $N_c = 5$. As explained in Section 3.3.3.3, an efficiency function \mathcal{E} must be introduced to retrieve the flame wrinkling lost with the thickening procedure. The model of Colin et al. (2000) is used here with the constant $\beta = 0.3$.

The part of the small scale turbulence filtered by the LES formalism is modeled using the subgrid-scale viscosity model WALE (Nicoud and Ducros (1999)).

7.2 Validation of the LES

We present, in the following, the results of the numerical simulations of flame acceleration in the BR30hS300 channel with adiabatic walls. LES results are validated against experimental data: 1) the flame tip velocity over the flame tip position; 2) overpressure time-traces from 7 transducers (their position is given in Section 6.1).

7.2.1 Flame propagation speed

The evolution of the flame tip velocity s_{tip} is displayed in Fig. 7.2. An overall good agreement with the experimental data (3 shots are provided) is observed. Upon ignition, the flame tip velocity is subsonic and approximately equal to $U_b = \Theta s_L = 3.22 \ m/s$ for an hydrogen/air flame at $\phi = 0.52$ and atmospheric conditions ($s_L = 0.59 \ m/s$ and $\Theta = 5.45$). The flame undergoes successive accelerations as it propagates in the obstructed channel. s_{tip} reaches a maximum s_{tip}^{max} just after the last obstacle. s_{tip}^{max} is very close to the sound speed at initial conditions on the burnt gas side $a_{b,0}$. s_{tip}^{max} is also approximately 250 times the initial flame speed, which shows that a combination of confinement and congestion in the flame path can provide very strong acceleration mechanisms. Note that strong fluctuations of the LES results around the experimental value can be observed in the obstructed region. Indeed, each obstacle provides an acceleration which is limited to its vicinity, as shown experimentally by Boeck et al. (2017b) who used a single fence-type obstacle, similar to the ones present in the BR30hS300 channel. The flame then slows down past the obstruction. The same behavior was also observed in the experiments of Boeck et al. (2017a) who used several ranges of fence-type obstacles. The surprisingly smooth experimental curves are the result of the low acquisition frequency. On average, the time of arrival of the flame is detected every 100 mm in the experiment, a distance higher than 8 times the obstacle streamwise thickness. This resolution does not allow to catch the aforementioned acceleration-deceleration phases.

After the last obstacle (at X = 2050 mm), there is no acceleration mechanism¹ able to

¹shock-flame constructive feedback, for example, can maintain high propagation speed provided that the degree of folding is lower than a critical value as shown in Deshaies and Joulin (1989)



Figure 7.2: Flame tip velocity s_{tip} as a function of the flame tip position X_{tip} . $a_{u,0}$ and $a_{b,0}$ are the sound speed at initial conditions of the reactants and products respectively. The vertical bars represent the position of the 7 obstacles. The numbers at the top of the figure indicate the location of the 7 pressure transducers. The corresponding overpressure signals are shown in Fig 7.3.

sustain these high flame propagation speeds, supersonic in the laboratory frame. Therefore, s_{tip} decreases during the flame propagation in the obstacle-free region to go below the sound speed of the reactants at initial conditions $a_{u,0}$. Approximately 4.2 m away from the ignition plate, a drastic decrease of the flame speed can be observed in the LES followed by a re-acceleration of the combustion wave. A strong planar shock propagating in the counter streamwise direction is responsible for this behavior. When the shock hits the flame, a stagnation of the latter is observed because of a backflow of fresh gases. The formation of this strong shock is also observed in the experiment, as discussed in Section 7.2.2.

7.2.2 Compressible effects

7.2.2.1 Overpressure generation in the BR30hS300 channel

Figure 7.3 displays the experimental and LES overpressure (i.e. $P - P_0$) records at probes 1 to 6. Note that the LES results are shifted in time by the same Δt_{record} . Δt_{record} is determined by matching the experimental time of arrival of the first pressure peak at the first probe. This procedure is necessary since the simulation starts with an already hemispherical flame, whereas, in the experiments, t = 0 s corresponds to the activation of the electric plug. This shift is commonly performed in the simulations of flame acceleration. The pressure signals are in very good agreement with the experimental data, except above $t \approx 0.065 \ s$. This last part of the signals is not relevant to the flame acceleration process. This is discussed in Section 7.3.

Figure 7.3 shows the high levels of overpressure that can be generated even without DDT



Figure 7.3: Overpressure signals for probes 1 to 6. Their position is indicated in Fig. 7.2 and is respectively $\{0.4, 1.4, 2.3, 3.2, 4.1, 5.0\}$ *m*. The symbols * and * indicate the time of arrival at different probes of a strong shock. The latter emerges few millimeters after the last obstacle and propagates downstream (*). It later reflects at the end plate and runs back upstream (*).

in the case of confined channels. The flame acts like a permeable piston which undergoes repeated accelerations in the obstructed region. This results in the generation of pressure waves and weak shocks ahead of the flame as discussed in Section 3.2.3 and observed at the first and second probes. A coalescence and steepening of these pressure waves later leads to the formation of a strong downstream running shock (*), which appears at the third probe, i.e. a few millimeters downstream of the last obstacle. This shock propagates in the channel with a quasi-constant speed around 932 m/s. This corresponds to a Mach number with respect to $a_{u,0}$: $Ma_s \approx 2.5$. It later reflects off the end plate and appears again in the sixth probe $(\underline{*})$. The discontinuity is now running upstream. It is detected in both the LES and the experiments. A good prediction of its arrival time at the different stations, thereby its Mach number, is observed. Taking into account the strength of this shock, one can conclude the deflagration must have at least slowed down when it crossed the shock's path in the experiment, similarly to what is predicted by the LES. The low acquisition frequency can explain why this is not visible in the experimental curves. The remaining part of the pressure signals corresponds to the same shock reflecting back and forth at the ignition and end plates.

The pressure signals, displayed in Fig. 7.3, are useful to validate the LES, however they do not allow to distinguish between two kinds of pressure waves/shocks: 1) the ones propagating ahead of the flame front, which can reflect off the obstacles and interact back with the flame; 2) those who journey in the hot gases far behind the flame front. The former type is of significant importance for the flame acceleration process since it can lead to flame/acoustic interactions and flame front instabilities (Richtmyer-Meshkov instability for example). Both are potential FA enhancers. The pressure signal prior to flame arrival is analyzed in the next section.



Figure 7.4: Zoom on the overpressure signals for probes 2 to 5. The signal starts when the flame interacts with the first obstacle and ends when the flame reaches the probe location. The evolution of the flame tip position X_{tip} is also displayed. A series of pressure peaks are formed when the flame is suddenly accelerated. The grey arrows associate the major pressure peaks to the obstacle responsible for their formation.

7.2.2.2 The flame/shock complex in obstructed channels

Figure 7.4 displays a zoom of the overpressure signals prior to flame arrival at probes 2 to 5. Therefore, it shows the overpressure generation ahead of the accelerating flame due to compressible effects. A few milliseconds after the flame interacts with a given obstacle, a peak of overpressure is observed at each probe. Consider the second probe first, located in the obstructed region. When the flame interacts with obstacle 2, the fresh gases are suddenly accelerated resulting in the formation of a series of compression waves which later coalesce and steepen to form a weak shock visible at $t \approx 0.036 \ s$. Successive shocks follow the first one and are attributed to the flame interaction with the subsequent obstructions. The third probe is placed 44 mm downstream of the last obstacle, thus it shows the shocks formed by the flame during its propagation in the obstructed region. The same pressure peaks observed in probe 2 are later detected by the third station, followed by those formed during the flame interaction with the subsequent obstacles. Probe 3 shows the capacity of repeated obstructions to produce high levels of overpressure (of the order of the bar) because of the piston effect. The BR30hS300 explosion illustrates the extreme damages that a deflagration can inflict to its surroundings when it propagates in an environment with high levels of confinement and congestion, even without transition to detonation.

The shocks formed at the last obstacle can catch up with the shocks generated at the previous obstacles for two reasons: 1) the fresh gases are repeatedly perturbed by the shock train which leads to a stepwise increase of the temperature of the reactants, hence the sound speed; 2) they are pushed by a faster flame. A comparison between the signals at the third, fourth and fifth stations shows how a strong shock forms by shock coalescence. It reaches probe 5 at $t \approx 0.042s$, where it is about to catch up with the leading shock.



Figure 7.5: Influence of the thermal treatment at the walls on the flame propagation speed (top) and the overpressure at probes 2, 3 and 5 (bottom). Only three stations are shown, but the same observations hold for the other probes.

All the shocks visible in Fig. 7.4 can be traced back to the flame propagation in the obstructed region. After their formation, they originally propagated in the region equipped with repeated obstacles where they have undergone a series of reflections at the obstacles to head back and cross the flame path. Shock/flame interaction can induce flame acceleration. However, we will see in Section 7.4.2 that it is not the main FA mechanism in the BR30hS300 channel.

7.3 Influence of the thermal boundary condition

In the previous section, the simulation results were obtained with adiabatic walls. Since the channel is entirely closed, heat fluxes at the walls may play an important role in the pressure build up in the chamber. To investigate the influence of the heat fluxes at the boundaries on the flame propagation, LES of the BR30hS300 channel with isothermal walls (wall temperature equal to 300 K) has been performed. Results are presented in Fig. 7.5. The flame propagation speed appears to be almost insensitive to the thermal treatment at the walls. s_{tip} at the exit of the obstructed section of the channel is slightly lower with the isothermal walls, but it remains within the range of experimental shots. Pressure signals for both simulations are also displayed at probes 2, 3 and 5 in Fig. 7.5 (bottom). They show that the signals prior to flame arrival are almost insensitive to changes in the heat fluxes at the walls. This means that the shocks, formed after the sudden accelerations of the flame, are almost identical for both cases, hence a comparable temperature increase of the mixture ahead of the flame. The combustion wave propagates in a mixture with similar properties which explains the comparable flame speeds observed in Fig. 7.5(top). Wall heat fluxes play a role a few milliseconds after the flame has passed each probe, i.e. in the hot gases far behind the reactive front. This region is too far from the deflagration front to impact the flame acceleration process. This inert part of the



Figure 7.6: Evolution of the mean pressure buildup in the channel $(\overline{P} - P_0)/(\overline{\gamma} - 1)$ as a function of the flame position for the isothermal case. The contribution of the heat release rate as well as the full RHS of Eq. 7.1 are also shown.

channel is filled with a mixture of combustion products, which are progressively relaxed to the isothermal walls temperature (300 K). Therefore, the reflected shocks propagate in a region with decreasing temperature and lower overall pressure compared to the adiabatic case. This leads to lower overpressure peaks.

One can grasp the impact of the wall heat fluxes by considering the evolution of the mean pressure inside the chamber. The channel being entirely closed, the main contributions to the mean pressure evolution are: 1) the heat release due to combustion; 2) the heat sink in the case of isothermal walls, which leads to Eq. 7.1 (the details of the derivation are presented in the Annex):

$$(\overline{P} - P_0)/(\overline{\gamma} - 1) = \int_0^t \overline{\omega}_T d\tau + \int_0^t \overline{\mathcal{Q}_w} d\tau$$
(7.1)

where $\overline{}$ denotes the mean over the channel volume. \mathcal{Q}_w denotes the heat fluxes at the walls. The mean pressure buildup inside the channel as a function of the flame position is displayed in Fig. 7.6. The contribution of the heat release rate as well as the full RHS of Eq. 7.1 are also shown. The RHS of Eq. 7.1 reproduces the pressure build up relatively well. Inside the obstructed region, the repeated increases in the flame surface leads to a stepwise build up of the mean pressure inside the channel. Until obstacle 4, the contribution of the wall heat fluxes remains marginal. Starting from obstacle 4, the volume occupied by the combustion products reaches approximately 21% of the volume of the channel and the role of the heat losses to the walls in the pressure budget becomes non-negligible. This explains the differences between the isothermal and the adiabatic LES in Fig. 7.5. Note that a sharp increase of the mean pressure is observed at $X_{tip} \approx 4500 \ mm$, which coincides with the interaction of the flame front with the reflected shock mentioned in Sections 7.2 and 7.2.2.

To conclude, the adiabatic walls assumption seems appropriate if one considers the flame acceleration only. However, reproducing the overpressure levels requires to take into account the heat losses to the walls. This is especially true in closed gas explosion chambers, but it was also confirmed in the case of vented channels (Volpiani et al. (2017a)).



Figure 7.7: Confinement induced flame acceleration following ignition. (Top) Flame speed s_{tip} as a function of the flame position X_{tip} given by the LES (black line) and Eq. (3.18) (green line). (Bottom) Flame surface \mathcal{A}_f as a function of the flame position X_{tip} given by the LES (black line). The blue dotted line corresponds to the surface of a sphere of radius X_{tip} . The numbers between parentheses indicate the flame tip position at which the 4 snapshots of Fig. 7.8 are taken.

7.4 Flame acceleration mechanisms involved in the BR30hS300 channel

Section 7.2.2 showed the capacity of a deflagration to produce high pressure levels when propagating in highly confined and obstructed configurations. The formation of strong shocks was traced back to the sudden accelerations of the flame front. The objective of this section is to identify the dominant mechanism behind these accelerations in the case of the BR30hS300 channel. Section 7.4.1 is dedicated to the few milliseconds following ignition, when the flame undergoes the first acceleration episode induced by the presence of the walls. Section 7.4.2 describes the processes by which obstacles impact the flame propagation.

7.4.1 FA before the first obstacle: effect of confinement

The BR30hS300 channel provides a strong confinement (very large length to height ratio 5400/60 = 90) along the flame path. The section of the channel downstream of the first obstacle is 250 mm long, and can be viewed as a narrow vented channel where the flame undergoes the first acceleration due to confinement. The flame speed s_{tip} and the flame surface \mathcal{A}_f as functions of the flame tip position X_{tip} are displayed in Fig. 7.7 for this region.

After initialization, the flame is hemispherical and propagates with the expected speed Θs_L . For a very short period of time, the flame remains spherical and no acceleration is observed. At $X_{tip} \approx 25 \ mm$, s_{tip} starts increasing and a deviation from the spherical shape

is observed. The acceleration continues until the flame skirt reaches the wall, which corresponds to the flame tip position X_{tip}^{wall} . Between $X_{tip} \approx 25 \ mm$ and X_{tip}^{wall} , s_{tip} increases linearly with X_{tip} , which corresponds to an exponential evolution of the flame speed with time. This behavior echoes the finger-flame effect discussed in Section 3.2.4. This is confirmed by the snapshot of the flame during this acceleration phase displayed in Fig. 7.8(1), where a finger shaped front is observed. The evolution of the flame speed is confronted to the theoretical formula Eq. (3.18) obtained by Bychkov et al. (2007). A fairly good agreement between theory and LES is observed. One can compare the corresponding acceleration rates σ_X during the finger flame propagation. σ_X is defined by $s_{tip} = \sigma_X X_{tip} + cste$. In Eq. (3.18), $\sigma_X = 2\alpha s_L \tanh(\mathcal{D}t)/(H/2)$, where $\alpha = \sqrt{\Theta(\Theta - 1)}$ and $\mathcal{D} = \alpha s_L/(H/2)$. As seen in Fig. 7.7, the theoretical curve quickly assumes its asymptotic linear behavior with a constant acceleration rate given by $\sigma_X = 2\alpha s_L/(H/2) = 193 \ s^{-1}$, which is lower than the one extracted from the LES $\sigma_X \approx 250 \ s^{-1}$.

The reason why the analytical expression is in fairly good agreement with the LES result is provided in Fig. 7.9 where the flow topology in the spherical and finger-shape regimes of propagation is displayed. First, it confirms the nature of the acceleration mechanism. The flame skirt moves towards the upper and bottom walls, pushing the hot gases towards the channel axis, where the flow deviates towards the direction of the flame tip propagation. This motion of burnt material pushes the flame front and is responsible for the acceleration. In addition, the flow in the hot gases appears rotational except in the two regions where the analytical expressions Eqs. (3.16- 3.17) are derived: near the ignition plate and close to the channel axis. The assumption of potential flow is confirmed in these two regions both during the spherical and finger shape regimes.

On the other hand, the differences between the LES and the theory can also be explained primarily by the fact that the BR30hS300 channel cross section is not square. In the theory, the problem is assumed axi-symmetric so that the confinement is the same in the Yand Z directions. In this channel however, the acceleration mechanism is not supported by the side walls, and the flame appears circular in the (X - Y) plane as highlighted in Fig. 7.8(2). For the same reason, the duration of the acceleration in the LES differs from the one predicted by Bychkov et al. (2007). Indeed, this mechanism is limited in time and stops as soon as the flame skirt touches the wall, which coincides with the time at which the flame tip position reaches the value $\Theta H/2$ according to Bychkov et al. (2007). This limit exceeds, nearly by a factor 2, the value X_{tip}^{wall} (see Fig. 7.7) obtained with the LES. Nevertheless, the rather good agreement between theoretical and LES acceleration rates indicates that the first acceleration undergone by the flame is primarily due to the confinement in the Z direction.

When the flame skirt touches the wall, a deceleration of the flame is observed. This is consistent with the numerical observations of Bychkov et al. (2007) and Valiev et al. (2013). It is worth noting that this deceleration is much less pronounced than in the aforementioned papers where square cross-section channels were considered. In Bychkov et al. (2007) and Valiev et al. (2013), the sudden slow down of the combustion wave corresponds to the moment when the flame skirt touches both the side walls and the top/bottom walls which terminates the finger flame acceleration process. In the BR30hS300 chamber, however, when the reactive front touches the top and bottom walls, the motion of the hot gases is still supported by the propagation of the flame skirt along the Y direction (l/H = 5). Similarly, the sharp flame surface decrease observed by Bychkov et al. (2007)² is balanced

²which is also confirmed in Fig. 7.8(1-2) where the flame shape in the (X - Z) plane goes from strongly elongated to slightly curved.



Figure 7.8: Snapshots of the flame at 4 positions indicated in Fig. 7.7. The isolevel of progress variable c = 0.5 is colored by the heat release rate H_{RR} . The numerical schlieren $\|\nabla\rho\|/\rho$ is visible on a (X-Z) cut through the channel axis (Y = 150mm). The cut is displayed on the channel wall so that it does not hinder the visualization of the flame shape.



Figure 7.9: Flow topology in the spherical (left) and finger shape (right) regimes. Arrows showing the velocity vector, colored by the temperature are superimposed to the heat release rate H_{RR} field.

here by the expansion of the front in the (X - Y) plane, so that \mathcal{A}_f stays roughly constant during this deceleration phase (see Fig. 7.7(bottom)).

Figure 7.7 shows that a second acceleration of the flame, exponential in time, occurs at $X_{tip} \approx 150 \ mm$ with $\sigma_X \approx 206 \ s^{-1}$. The same mechanism discussed above, but induced by the side walls this time, could be responsible for this second acceleration as suggested by the elongated cylindrical shape of the front as the flame skirt approaches the side walls in Fig. 7.8(3-4). The flame appears almost planar in the Z direction so that the effect of the side wall can be considered essentially two-dimensional. Valiev et al. (2013) predict the following expression for σ_X in the 2D case: $\sigma_X = (\Theta - 1)s_L/(l/2) = 15.3 \ s^{-1}$. Therefore, this mechanism alone can not explain the observed increase of the flame speed. However, a perturbation of the flame front is also triggered at $X_{tip} = 150 \ mm$, and coincides with the inversion of the flame skirt shown in Fig. 7.8(3). The growth rate of this perturbation combined with side walls effect could explain the acceleration. It is important to note that the emergence of the perturbation of the flame front in the LES must be taken with caution since it can not be confirmed by the few experimental data available.

7.4.2 Flame/obstacle interaction as FA enhancer

The previous section showed that confinement alone can provide strong acceleration mechanisms. In the region comprised between the ignition plate and the first obstacle, the flame speed has been multiplied by nearly a factor 8. Obstacles are also known to enhance flame acceleration. In the present channel, the flame speed increased in the obstructed region from $\approx 22m/s$ to $\approx 810m/s$, i.e. by nearly a factor 37. We will discuss, in the following, the different mechanisms contributing to this strong acceleration. First, global observations on the impact of the obstacles on the deflagration front are presented in Section



Figure 7.10: Flame interaction with the first obstacle. The isolevel of progress variable c = 0.5 is colored by the heat release rate H_{RR} . The numerical schlieren $\|\nabla\rho\|/\rho$ is visible on a (X-Z) cut through the channel axis (Y = 150mm). The cut is displayed on the channel wall so that it does not hinder the visualization of the flame shape.

7.4.2.1. Then, the relative importance of flow contraction and turbulence to the flame acceleration mechanism is discussed in Section 7.4.2.2.

7.4.2.1 Flame front interaction with repeated obstacles

As the flame approaches the first obstacle, its front is strongly elongated, because of the side walls effect, and appears quasi-planar in the vertical direction. Because of the hight front curvature in the X - Y plane, the effect of the obstruction is not felt uniformly along the reactive surface. The most advanced points on the flame are the first to be drifted by the flow and form a cylindrical head as they penetrate the region between the top and bottom fences. This is illustrated in Fig. 7.10. In the bottom snapshot, when the flame tip exits the obstacle, the reactive surface has drastically increased: the flame skirt has not reached the side wall yet and is still attached to the ignition plate (i.e. $X_{skirt} = 0$), whereas the flame tip has reached the position $X_{tip} \approx 400 \ mm$.

The flame journey in the first obstruction also impacts the fresh gases far ahead. The latter are suddenly accelerated by the motion of the flame, promoting vortex shedding downstream of the second obstacle. Consequently, starting from obstacle 2, the effect of the obstacles on the flame will no longer be restricted to the vicinity of the fences but will extend to a certain distance downstream. This is highlighted in Fig. 7.11, where the flame exits the obstruction with noticeable folding due to its interaction with the flow structures. Note that relatively large pockets of fresh gases are still trapped in the region close to the side walls and upstream of the first obstacle, contributing to the overall energy release in the closed chamber.



Figure 7.11: Flame interaction with the second obstacle. The isolevel of progress variable c = 0.5 is colored by the heat release rate H_{RR} . The numerical schlieren $||\nabla \rho||/\rho$ is visible on a (X - Z) cut through the channel axis (Y = 150mm). The cut is displayed on the channel wall so that it does not hinder the visualization of the flame shape.

The flame interaction with the third and fourth obstacles is displayed in Fig. 7.12. A clear transition to a turbulent regime of propagation is shown. Because of the strong surface folding, the burning rate increases. Consequently, the pockets of fresh material trapped in the corners upstream of the obstacles, which can be seen in Fig. 7.11(2) for example, are consumed more efficiently. The flame skirt can then catch up with the rest of the flame a few microseconds after the flame tip exits the fence. Therefore, between two successive obstacles, the flame propagates as a strongly wrinkled flamelet with a low overall curvature in the (X - Y) plane. The first three snapshots of Fig. 7.12 also show how the turbulent fluctuations behind obstacle 4 intensify as the flame approaches.

7.4.2.2 Evolution of the flow downstream of the obstacles

Figures 7.8-7.12 provide a global description of the flame journey in the obstructed region. They show the strong constructive feedback between the accelerating flame and the flow ahead of it. This triggers a combination of FA mechanisms that still need to the be identified. To tackle the obstacle induced flame acceleration problem, it is mandatory to identify the ingredients controlling the flame propagation. Any given point X_f of the flame surface propagates for two reasons: 1) it is drifted by the flow ahead of it; 2) it consumes the fresh gases with normal velocity. The evolution of X_f is given by the



Figure 7.12: Flame interaction with the third and fourth obstacles. The isolevel of progress variable c = 0.5 is colored by the heat release rate H_{RR} . The numerical schlieren $||\nabla \rho||/\rho$ is visible on a (X - Z) cut through the channel axis (Y = 150mm). The cut is displayed on the channel wall so that it does not hinder the visualization of the flame shape.

following equation written with respect to the fresh gases:

$$\frac{d}{dt}X_f(Y,Z;t) = u_u(Y,Z;t) + \mathcal{E}_{fold}(Y,Z;t)s_L$$
(7.2)

where u_u is the flow velocity and $\mathcal{E}_{fold}s_L$ is the local turbulent burning velocity, with \mathcal{E}_{fold} accounting for the increase of the flame surface. Consequently, explaining the strong FA process occurring in the BR30hS300 channel reduces to identifying the mechanisms that can trigger a strong increase of either one or both terms in Eq. (7.2). To do so, the flow ahead of the flame is inspected in the following.

Velocity and vorticity fields are displayed in Fig. 7.13. The velocity is normalized by the absolute speed s_{tip}^n of the flame as it approaches obstacle n. The picture shows how turbulent fluctuations emerge and intensify in the unburnt gas flow. It also suggests that a combination of two ingredients is responsible for the strong flame acceleration observed in the BR30hS300 channel.

Flame-Vortex interactions (FVI)

Spots of rotational motion become noticeable in Fig. 7.13(right) downstream of obstacle 2 when the flame approaches. Frame 2 shows that the impact of the moving flame is not restricted to the obstruction just ahead of it but extends to the following obstacle: vortex shedding appears downstream of obstacle 3. Two important features are worth mentioning regarding frames 1-2: 1) the flow keeps its initial symmetry, which implies that the flame itself will remain symmetric 3 ; 2) the flow along the channel horizontal centerline

³This is confirmed in Fig. (7.10 - 7.11).



Figure 7.13: Cuts through the channel axis describing the flow as the flame approaches the successive obstacles. The isocontour of the progress variable c = 0.5, in orange, shows the flame front. (Left) Nondimensional velocity magnitude. s_{tip}^n is the flame speed just prior to obstacle n. (Right) Magnitude of the vorticity.



Figure 7.14: Resolved \mathcal{A}_{f}^{res} and total \mathcal{A}_{f}^{tot} flame surfaces as a function of the flame tip position X_{tip} . \mathcal{A}_{f}^{tot} includes the contribution of the efficiency model.

is essentially irrotational, which implies that the flame tip will be drifted by the flow along the channel axis while the other points on the reactive surface will interact with either the recirculation zone in frame 1 or the vortices in frame 2. These two observations explain the elongated shape of the flame observed at the exit of obstacles 1 and 2 (see Figs. 7.10-7.11). The symmetry breakdown occurs when the flame is close to obstacle 3.

As the flame propagates in the channel, an increase of the vorticity level is observed. The strong mixing zones also extend to a greater distance downstream of the obstacles. This leads to enhanced Flame/Vortex Interactions (FVI). The subsequent wrinkling of the flame front contributes to the second term in the RHS of Eq. (7.2). The importance of FVI for the flame acceleration can be grasped by looking at the evolution of the flame surface as a function of the flame tip position. In Fig. 7.14, both the resolved \mathcal{A}_{f}^{res} and total \mathcal{A}_{f}^{tot} flame surfaces are displayed. \mathcal{A}_{f}^{res} corresponds to the flame surface resolved on the computational grid, whereas \mathcal{A}_{f}^{tot} includes the contribution of the Colin et al. (2000) efficiency model, designed to retrieve the lost subgrid scale wrinkling due to the artificial thickening of the flame. Up to 700 mm, a globally monotonic evolution of \mathcal{A}_{f}^{res} is observed because of the strongly elongated aspect of the flame as discussed in Section 7.4.2.1. Pockets of fresh material left behind the flame are then progressively consumed so that \mathcal{A}_{f}^{res} starts decreasing when the flame approaches obstacle 3. Starting from obstacle 3, the same behavior is observed between two obstacles and can be summarized as follows: 1) \mathcal{A}_{f}^{res} drastically increases at the exit of the obstruction due to flame-vortex interactions; 2) when the flame exits the strong mixing zone and the flame skirt catches up with the flame tip, \mathcal{A}_{f}^{res} decreases. Note that shocks, evidenced in Section 7.2.2, also contribute to the flame surface increase when they reflect off the obstacles and interact back with the flame.

The evolution of \mathcal{A}_{f}^{tot} is very similar to \mathcal{A}_{f}^{res} . It shows a strong contribution of the efficiency model to the turbulent burning velocity: on average, the total flame surface is approximately two times the resolved surface. It is worth noting that the activation of the Colin et al. (2000) efficiency model before the second obstacle was not expected. It is certainly due to an overestimation of the velocity fluctuations in the channel by the operator Eq. 3.32 and the constant wrinkling formulation adopted in the present study. Indeed, the equilibrium assumption between turbulence and flame wrinkling is certainly not valid in this area. A dynamic model, as the one used by Volpiani et al. (2017a), would probably be more appropriate to handle this transition to turbulence.

FVI appears as a good candidate to explain the flame acceleration process in the BR30hS300



Figure 7.15: Flame propagation speed s_{tip} (solid line) and total flame surface \mathcal{A}_f^{tot} (dashed line) as a function of the flame tip position X_{tip} . FVI and FC stand for Flame-Vortex Interaction and Flow Contraction respectively. Gray areas correspond to regions of strong flame deceleration. Areas inside the transparent boxes correspond to moderate deceleration or almost constant velocity.

channel. The total flame surface is displayed along with the flame propagation speed in Fig. 7.15. We are interested in the region comprised between the second obstacle and $X_{tip} \approx 2500 \ mm$, where FVI occurs downstream of each obstruction. It is seen that the increase of the flame surface, just downstream of each obstacle, does not correspond to flame acceleration episodes. The FA mechanism seems to be limited to the vicinity of the obstacles. The regions where \mathcal{A}_{f}^{tot} increases coincide with regions of negative slope in the s_{tip} curve. This clearly rules out FVI from being the main acceleration mechanism in the present case. However, it still plays an important role: it helps maintain high propagation velocities in the regions comprised between two obstructions. Consider one of these regions. The flame first slows down when it exits the congestion (gray area) before reaching an almost constant velocity (or very moderate deceleration). This plateau (transparent box) coincides with the increase of the flame surface: in these zones, FVI intensifies flame folding and increases the flame burning velocity. As soon as the flame surface starts decreasing (prior to the next obstacle), a drastic decrease of the flame speed is observed (second gray area), which is attributed to the lost contribution of flame folding to the burning velocity caused by the flow "relaminarization". Indeed, the vortex shedding effect, downstream of an obstacle, is limited to a distance lower than the large spacing separating two consecutive obstacles as shown in Fig. 7.13(right).

Sudden Flow Contraction (FC)

Figure 7.15 shows that the turbulence generated by the obstacles is not responsible for accelerating the flame. The obstructions involve another FA mechanism that needs to be identified. The answer, as mentioned in Fig. 7.15, lies in the sudden Flow Contraction (FC), a mechanisms able to increase both terms in the RHS of Eq. (7.2): 1) in Fig. 7.13, the fresh material, pushed by the flame, is forced to accelerate through the fence to attain



Figure 7.16: Nondimensional flame propagation speed $(s_{tip} - s_{tip}^n)/s_L$ as a function of the nondimensional flame tip position $(X_{tip} - X_{Obs}^n)/H_{free}$ arount each obstacle n. s_{tip}^n is the flame speed at the center of obstacle n. $H_{free} = (1 - \varsigma)(H/2)$ is the free space between the top and bottom obstructions. X_{Obs}^n is the position of obstacle n.

velocities higher than the flame propagation speed; 2) as the flame head penetrates the congestion, it stretches because of the velocity gradient ahead of it, thereby increasing the \mathcal{E}_{fold} term. As a result, FC is the dominant FA mechanism in the BR30hS300 channel. The flow around the obstacles also explains why FA is limited to the vicinity of the fences, since the Flow Expansion (FE), at the exit of the obstacle, has the opposite effect (to FC) and results in the decrease of the flow velocity a certain distance downstream. This distance depends strongly on the flame speed when approaching the obstacle.

The capacity of repeated FCs to produce very fast flames is highlighted in Fig. 7.16. Each time the flame goes through a flow contraction, the flame speed increases quasi-linearly with the flame position. This corresponds to an exponential increase of s_{tip} in time, hence a strong FA mechanism. Moreover, even though all the obstacles provide the same blockage ratio, they do not induce the same flame acceleration rate. The nondimensional acceleration rate $\sigma_{Obs}^n = \sigma_X H_{free}/s_L$, where $H_{free} = (1 - \varsigma)(H/2)$, is evaluated and displayed for each obstacle in Fig. 7.17. σ_{Obs}^n strongly increases with increasing n. This can be explained by the fact that the effect of FC depends on the velocity at which the fresh material impacts the contraction, which in turn depends on the speed at which the flame approaches the obstacle. Since the flame reaches each obstruction with increasing velocity, the effect of FC on the flame is also stronger with increasing n. The evolution of σ_{Obs}^n with the flame speed is displayed in Fig. 7.17(right). The flame speed is taken close to the obstacle, when the obstacle induced acceleration starts (i.e. when the slope of the flame speed curve becomes close to σ_{Obs}^n) and is denoted $\mathcal{U}_{f\to Obs}^n$. $\mathcal{U}_{f\to Obs}^n$ is also the velocity at which the fresh material, pushed by the flame, impacts the fence. Note that obstacle 5 exhibits a singular behavior, which is not explained.



Figure 7.17: Evolution of the nondimensional flame acceleration rate $\sigma_{Obs}^n = \sigma_X H_{free}/s_L$ (left) as a function of the obstacle number n and (right) as a function of the speed s_{tip} when the flame is close to the obstacle (i.e. when the slope of the s_{tip} curve reaches σ_{Obs}^n). This speed is denoted $\mathcal{U}_{f\to Obs}^n$ and can be viewed as the velocity at which the fresh material, pushed by the flame, impacts the fence.

7.4.3 Linking the flame acceleration rate to the flow contraction

In the following, an attempt to directly link the flame acceleration rate through an obstruction σ_{Obs}^n to the flow contraction is presented. The objective is to highlight the laminar nature of this mechanism and the ingredients by which it induces flame acceleration. This analysis is restricted to the laminar regime, which mainly concerns obstacles 1 and 2. It is inspired from the paper of Bychkov et al. (2005) on accelerating flames in narrow smooth tubes. Indeed, once the flame tip penetrates the obstruction and until it exits the latter, the system is very similar to the one considered in Bychkov et al. (2005); the only difference lies in the initial conditions, so that the solution remains similar except for the formula for the accelerating term.

Consider a planar flame propagating in a channel of height H reaching a sudden contraction ($\varsigma = 2h/H = 30\%$) with the non-dimensional speed $\vartheta = U_{f \to Obs}^n/s_L$ in Fig. 7.18. The coordinates of the system are normalized by H_{free} : $(\xi, \eta) = (X, Z)/H_{free}$, since we are interested in the flame journey in this congestion. A nondimensional time is introduced: $\tau = ts_L/H_{free}$. The flow velocity in the unreacted material ahead of the flame is also normalized: $\vartheta_u = (u_X, u_Z)/s_L$. As shown in Fig. 7.17, the flame accelerates exponentially in time during its journey in the obstructed area, so that the evolution of s_{tip} takes the form:

$$s_{tip}/s_L \propto \exp(\sigma_{Obs}^n \tau)$$

and σ_{Obs}^n is an eigenvalue of the problem.

As shown in Fig. 7.18, the problem is further simplified by neglecting the flow separation that occurs just downstream of the sudden contraction. Moreover, we assume that the fresh gases, inside the congestion, are pushed by a plane-parallel flow. The governing equation for such a flow is:

$$\frac{\partial \vartheta_{u,X}}{\partial \tau} = -\frac{\partial p}{\partial \xi} + \frac{1}{Re_f} \frac{\partial^2 \vartheta_{u,X}}{\partial \eta^2}$$
(7.3)



Figure 7.18: A planar flame approaching a sudden contraction at speed $\mathcal{U}_{f\to Obs}^n$. The problem is simplified by neglecting the flow separation.

where the density and pressure are scaled by ρ_u and $\rho_u s_L^2$ respectively. $Re_f = s_L H_{free} \rho_u / \mu$ is the flame Reynolds number and is equal to 661.1 for the present flame. The pressure gradient is a function of time only and is the key ingredient for the flame acceleration. Contrary to Bychkov et al. (2005), it is triggered, in the present case, by the pressure loss at the contraction and sustained by the flame motion. During the regime of propagation considered here, the flame speed remains subsonic and we assume that during the brief flame interaction with the obstacle no weak shock is formed so that the pressure gradient is proportional to the increase of the burning rate. Therefore, the pressure gradient takes the form: $-\frac{\partial p}{\partial \xi} = \Lambda_1 exp(\sigma_{Obs}^n \tau)$.

Since the flame pushes the fresh gases with an exponentially increasing speed, the flow velocity in the reactants takes the form: $\vartheta_{u,X} = \varphi(\eta) \exp(\sigma_{Obs}^n \tau)$. Finally, Eq. (7.3) becomes:

$$\beta_f^2 \varphi = \Lambda_2 + \varphi'' \tag{7.4}$$

where $\beta_f = \sqrt{\sigma_{Obs}^n Re_f}$. Imposing a no-slip boundary condition at the walls $\varphi(\pm 1) = 0$, the solution to Eq. (7.4) reads:

$$\varphi = \Lambda_3 \frac{\cosh(\beta_f) - \cosh(\eta\beta_f)}{\cosh(\beta_f) - 1} \tag{7.5}$$

Therefore, the velocity profile generated by the accelerating piston reads:

$$\vartheta_{u,X} = \Lambda_3 \exp(\sigma_{Obs}^n \tau) \frac{\cosh(\beta_f) - \cosh(\eta\beta_f)}{\cosh(\beta_f) - 1}$$
(7.6)

To determine an expression for Λ_3 , we assume that before the flame enters the congested zone, the fresh gases trapped between the reactive front and the obstacle are pushed at the piston speed ϑ . Therefore, because of the contraction, the average flow velocity at the entry of the obstruction takes the form: $\langle \vartheta_{u,X}(\eta,0) \rangle = \Psi(\varsigma)\vartheta$, where $\Psi(\varsigma)$ is a contraction coefficient depending on the blockage ratio ς . During its journey through the obstruction, the flame quickly deviates from the planar shape, with the distortion factor $\Omega_f(\tau)$. This results in the evolution of the average velocity estimated by: $\langle \vartheta_{u,X}(\eta,\tau) \rangle \approx$ $\Psi(\varsigma)\vartheta\Omega_f(\tau)$. Then, the velocity profile reads:

$$\vartheta_{u,X} = \Psi(\varsigma)\vartheta\Omega_f(\tau)\frac{\cosh(\beta_f) - \cosh(\eta\beta_f)}{\cosh(\beta_f) - \beta_f^{-1}\sinh(\beta_f)}$$
(7.7)

The velocity profile given by Eq. (7.7) is displayed in Fig. 7.19 for $Re_f = 661.1$ (present



Figure 7.19: Velocity profile $\vartheta_{u,X}$ inside the contraction obtained with Eq. (7.7) normalized by its maximum value. The Reynolds number for the present flame is: $Re_f = 661.1$. Profiles are shown for different values of the acceleration rate $\sigma_{Obs}^n \tau = 0.01, 0.1, 1, 10$.

flame) and different values of $\sigma_{Obs}^n \tau = 0.01, 0.1, 1, 10$. It shows that, for an infinitely slow acceleration of the flame, the velocity profile tends to a Poiseuille-like flow, whereas for large values of $\sigma_{Obs}^n \tau$, the flow is composed of two boundary layers and a main stream where the velocity is uniform. This is confirmed in Fig. 7.20, where the velocity profiles obtained numerically for obstacles 1, 2 and 3 are compared to the analytical expression. A quasi uniform main stream is observed, which is consistent with Eq. (7.7). The main difference between the analytical expression and LES result is the presence of the recirculation zone near the walls which is neglected in the present theory. Nevertheless, the comparison is satisfactory and validates Eq. (7.7).

The remaining pieces of the derivation follow the steps taken by Bychkov et al. (2005). Now that we have an expression for the velocity of the fresh material pushed by the flame, we know how each point on the combustion wave is drifted by the flow. The flame shape is defined with respect to the flame tip: $\xi_f(\eta, \tau) = \xi_{tip}(\tau) + \mathcal{C}(\eta, \tau)$. The flame can be supposed planar in the channel axis so that the evolution of ξ_{tip} is given by Eq.(7.8). However, away from the channel axis, the fuel consumption rate increases by a factor equal to the increase of the flame surface relative to a planar flame. Therefore we have:

$$\frac{\partial \xi_{tip}}{\partial \tau} = \vartheta_{u,X}(0,\tau) + 1 \tag{7.8}$$

$$\frac{\partial \xi_f}{\partial \tau} = \vartheta_{u,X}(\eta,\tau) + \left[1 + \left(\frac{\partial \mathcal{C}}{\partial \eta}\right)^2\right]^{1/2}$$
(7.9)

We then obtain an equation for the evolution of \mathcal{C} :

$$-\frac{\partial \mathcal{C}}{\partial \tau} = \vartheta_{u,X}(0,\tau) - \vartheta_{u,X}(\eta,\tau) + 1 - \left[1 + \left(\frac{\partial \mathcal{C}}{\partial \eta}\right)^2\right]^{1/2}$$
(7.10)

Equation (7.10) is linearized by assuming that $\left(\frac{\partial \mathcal{C}}{\partial \eta}\right)^2 \gg 1$. Moreover, taking into account the symmetry of the problem, only the part $\eta > 0$ is considered in the following and



Figure 7.20: Validation of the velocity profile obtained with the analytical expression Eq. (7.7) (solid line) for $\sigma_{Obs}^n \tau = 20$. The dashed lines correspond to the velocity profile extracted from the cuts of the velocity field at Y = 150 mm.

Eq. (7.10) becomes:

$$-\frac{\partial \mathcal{C}}{\partial \tau} = \vartheta_{u,X}(0,\tau) - \vartheta_{u,X}(\eta,\tau) + \frac{\partial \mathcal{C}}{\partial \eta}$$
(7.11)

with a solution taking the form: $C(\eta, \tau) = \Phi(\eta) \exp(\sigma_{Obs}^n \tau)$. For $\eta > 0$, we also have:

$$\Omega_f(\tau) = \int_0^1 \left[1 + \left(\frac{\partial \mathcal{C}}{\partial \eta} \right)^2 \right]^{1/2} d\eta$$
$$= -\mathcal{C}(1,\tau)$$

Finally, the evolution of the flame shape Φ follows the equation:

$$\Phi' = -\sigma_{Obs}^{n} \Phi + \Psi(\varsigma) \vartheta \Phi(1) \frac{\cosh(\eta \beta_f) - 1}{\cosh(\beta_f) - \beta_f^{-1} \sinh(\beta_f)}$$
(7.12)

Integrating Eq. (7.12) gives:

$$\Phi(\eta) = \frac{\Psi(\varsigma)\vartheta\Phi(1)\exp(-\sigma_{Obs}^n\tau)}{\cosh(\beta_f) - \beta_f^{-1}\sinh(\beta_f)} \int_0^\eta (\cosh(x\beta_f) - 1)\exp(-\sigma_{Obs}^n x)dx$$
(7.13)

Taking $\eta = 1$, one can get rid of the term $\Phi(1)$. Since in our case $\sigma_{Obs}^n > 20$, we have $\beta_f \gg 1$ and Eq.(7.13) gives a formula for the acceleration rate:

$$\sigma_{Obs}^{n} = \frac{(Re_f - 1)^2}{4Re_f} \left(\sqrt{1 + \frac{4Re_f(\Psi(\varsigma)\vartheta + 1)}{(Re_f - 1)^2}} - 1 \right)^2$$
(7.14)

Equation (7.14) can be further simplified in the case of large Reynolds number $Re_f \gg 4\Theta$, which leads to the following expression:

$$\sigma_{Obs}^{n} = \frac{(\Psi(\varsigma)\vartheta + 1)^2}{Re_f}$$
(7.15)

Equation (7.15) predicts that:

- σ_{Obs}^{n} increases with increasing blockage ratio ς , other parameters being fixed. This is consistent with the observations of Johansen and Ciccarelli (2013), who investigated the effect of congestion ($\varsigma = 0.3, 0.5, 0.67$) on the flame propagation. The experiment consists of an entirely closed square channel (cross-section 0.076×0.076) equipped with a series of fence-type obstacles. The chamber is filled with a stoichiometric mixture of methane/air at 47kPa. The results are displayed for the first obstacle in Fig. 7.21. They show that even though the flame approaches the obstacles with approximately the same speed, its acceleration rate through the obstruction increases with increasing ς . The same behavior was observed by Masri et al. (2000) using other types of obstacles and a Liquefied Petroleum Gas (LPG) (88% C₃H₈, 10% C₃H₆ and 2% C₄H₁₀ by vol.).
- σ_{Obs}^n decreases with increasing flame Reynolds number, for fixed flame speed ϑ and contraction coefficient $\Psi(\varsigma)$. Further numerical simulations are currently being performed to assess this point.

Contrary to other flame acceleration mechanisms (finger flame, tulip flame, DL ...), the acceleration rate associated with sudden obstructions does not depend on the flame only, but is strongly linked to the speed at which the latter impacts the congestion. This makes the process more powerful when coupled to other obstructions, even without an additional contribution of turbulence. Eq. (7.15) still needs an expression for the contraction coefficient $\Psi(\varsigma)$ to be confronted to the LES. Let us consider obstacle 1 first. Figure 7.10 shows that the head of the flame forms a cylindrical front as it goes through the congestion. Therefore, even though the channel is of rectangular section, the problem can be considered as locally cylindrical so that an expression for $\Psi(\varsigma)$ can be obtained from the square of the upstream to the downstream channel radius ratio: $\Psi(\varsigma) = 1/(1-\varsigma)^2$. $\sigma_{Obs}^1 = 11.77$, which is about half the value given by the LES. However, the presence of the flow separation, highlighted in Fig.7.20 and visible in Fig.7.13, increases the effective $\frac{1}{2}$ blockage ratio of the obstruction ς_2 . ς_2 can be deduced from the velocity profiles given by the LES by subtracting the width of the recirculation zone from H_{free} . Doing so we have $\varsigma_2 \approx 0.37$, which gives $\sigma_{Obs}^1 = 19.84$ in reasonable agreement with the value extracted from the LES: $\sigma_{Obs}^1 \approx 22$. Since the flow, before the flame hits obstacle 2, is still laminar, we could have expected a good agreement with the LES for the second congestion as well. However, the analytical expression reproduces σ_{Obs}^2 with a 20% accuracy only: 104.22 and 84 for the theory and the LES respectively.

Finally, we would like to point out that Eq. (7.15) can not be considered satisfactory since a formula for $\Psi(\varsigma)$ is still missing. Further derivations are necessary if one wants to obtain a consistent analytical expression for the flame acceleration rate σ_{Obs} .

Annex

For a reactive flow, the equation of conservation of the sensible energy e_s reads:

$$\rho \frac{De_s}{Dt} = \dot{\omega}_T + \frac{\partial}{\partial x_i} (\lambda \frac{\partial T}{\partial x_i}) - \frac{\partial}{\partial x_i} \left(\rho \sum_{k=1}^N h_{s,k} Y_K V_{k,i} \right) + \sigma_{ij} \frac{\partial u_i}{\partial x_j} + f_{vol}$$
(7.16)

Suppose that C_p and C_v are temperature invariant. The sensible energy can be linked to the pressure via:

$$\rho e_s = P/(\gamma - 1) - \rho C_p T_0$$



Figure 7.21: Experimental data of flame acceleration through sudden contraction extracted from Johansen and Ciccarelli (2013). They show that the acceleration rate of a stoichiometric methane/air flame, initially at 47kPa, through the first obstacle depends on the congestion provided by the obstacle ($\varsigma = 0.3, 0.5, 0.67$).

Using the mass conservation equation, the LHS of Eq. 7.16 becomes:

$$\rho \frac{De_s}{Dt} = \frac{\partial}{\partial t} \left(P/(\gamma - 1) \right) + \frac{\partial}{\partial x_i} \left(u_i P/(\gamma - 1) \right)$$
(7.17)

which allows to retrieve the following equation for the pressure, neglecting the contributions of the viscous tensor and the external volumic forces:

$$\frac{\partial}{\partial t} \left(P/(\gamma - 1) \right) = \dot{\omega}_T + \frac{\partial}{\partial x_i} \left(\lambda \frac{\partial T}{\partial x_i} \right) - \frac{\partial}{\partial x_i} \left(u_i P/(\gamma - 1) \right)$$
(7.18)

suppose that the integral and derivative operators commute and that the pressure is homogeneous in the chamber, an equation for the evolution of the mean pressure can be obtained:

$$\frac{d}{dt}\left(\overline{P}/(\gamma-1)\right) = \overline{\dot{\omega}_T} + \overline{\mathcal{Q}_w} - \iiint_{\mathcal{V}} \frac{\partial}{\partial x_i} \left(u_i P/(\gamma-1)\right) d\mathcal{V}/\mathcal{V}$$
(7.19)

$$\frac{d}{dt}\left(\overline{P}/(\gamma-1)\right) = \overline{\dot{\omega}_T} + \overline{\mathcal{Q}_w} - \iint_{\mathcal{S}} \left(P/(\gamma-1)\right) \overrightarrow{u} \cdot \overrightarrow{d\mathcal{S}}/\mathcal{V}$$
(7.20)

$$\frac{d}{dt}\left(\overline{P}/(\gamma-1)\right) = \overline{\dot{\omega}_T} + \overline{\mathcal{Q}_w} \tag{7.21}$$

where we have taken into account the zero normal velocity at the walls.

Chapter 8

The role of shocks in the spontaneous initiation of detonations

Abstract

In Chapter 6, two hydrogen/air explosion scenarios were selected from the BR30hS300 database: a very lean mixture ($\phi = 0.52$) where only flame acceleration was observed and the stoichiometric case where DDT was triggered. Chapter 7 was dedicated to the numerical investigation of the deflagrating case. It showed that a combination of powerful FA mechanisms was responsible for the supersonic regime of propagation achieved by the flame inside the obstructed region of the channel. Ahead of the combustion wave, the formation of a strong shock was highlighted and traced back to the sudden accelerations of the flame at the obstacle locations. Such discontinuities are known to trigger DDT Oran and Gamezo (2007), thereby it is important to understand why, in this case, DDT was not observed to assess the necessary conditions that must be satisfied to trigger such a violent transition. This will prepare the ground for the numerical study of the stoichiometric case performed in Chapter 9.

The commonly recognized mechanism for DDT is first described in Section 8.1. It was predicted by Zeldovich et al. (1970); Zeldovich (1980) and involves the preconditioning of a small pocket of reactants by a gradient of reactivity. In Section 8.2, the lean case $\phi = 0.52$ discussed in Chapter 7 is analyzed with regards to DDT to show that the necessary conditions for ignition are not met. An overview of different mechanisms that can explain the onset of detonation in the case $\phi = 1$ is finally provided in Section 8.3.

8.1 Spontaneous initiation of detonations

There is a general consensus that a detonation front can emerge from a local thermal explosion ahead of a very fast flame. The process, known as the Zeldovich's mechanism, involves the sudden ignition of a pocket of unburned material. Since the reactants are initially at a frozen state, the reactants must be heated, by some source generated during the flame propagation, to escape the frozen state. We assume here that a pocket of unburned material has been brought to an ignitable state (T_u, P_u) . The spontaneous initiation of a detonation from this hot spot is explained in Section 8.1.2.



Figure 8.1: Sketch of temperature, pressure and heat release history during the constant volume ignition of a reactive mixture initially at (T_u, P_u) . $T_{b,I}$ and $P_{b,I}$ are the equilibrium temperature and pressure respectively.

8.1.1 Ignition characteristic times

The spontaneous detonation initiation mechanism involves ignition processes characterized by different time scales. We define here characteristic times that can be distinguished during the ignition of a fixed volume of reactive material. Figure 8.1 displays a typical temperature, pressure and heat release history during the ignition of the mixture. Bradley and Kalghatgi (2009); Bates et al. (2016); Bates and Bradley (2017) define two ignition characteristic times based on the profile of H_{RR} :

- the induction time τ_I , defined as the time necessary for a mixture at certain conditions of temperature and pressure to reach the maximum H_{RR} ;
- the excitation time τ_E , defined as the duration of the chemical heat release during ignition. It is evaluated as the time necessary for the heat release rate to go from 5% to 100% its maximum value. τ_E can be two or three orders of magnitudes smaller than τ_I .

The set of parameters (τ_I, τ_E) depends on the mixture and on the initial temperature and pressure conditions. During the constant volume ignition, both temperature and pressure increase to reach the burnt gas state $(T_{b,I}, P_{b,I})$.

8.1.2 Requirements to generate a self-propagating CJ wave

The structure of a detonation front was discussed in Chapter 4. It involves a strong leading shock that raises the temperature of the reactants to the point (called VN state) where chemical reactions can be initiated. One can already postulate a necessary condition to generate a self-propagating CJ wave from a pocket of unburned material: pressure must increase rapidly to reach values at least equal to $P_{N_{CJ}}$. Let us see if this is possible from the combustion of a constant volume pocket of reactants. At equilibrium, the gas pressure is given by $P_{b,I} = (T_{b,I}/T_u)P_u$, so that:

$$P_{b,I}/P_{N_{CJ}} = \frac{(T_{b,I}/T_u)P_u}{P_{N_{CJ}}} = \frac{T_{b,I}^{<15}}{P_{N_{CJ}}/P_u} < 1/2$$



Figure 8.2: (Left) Sequential ignition of successive slices of gas inside a region of nonhomogeneous induction time (hot spot). (Right) Instantaneous temperature and pressure profiles inside the hot spot illustrating the formation of the induction front and the compression wave propagating respectively at speed u_I and a.

This already rules out the possibility of detonation initiation from the combustion of a constant volume pocket of reactants (Clavin and Searby (2016)).

A way of creating a sufficiently high and rapid compression inside a pocket of reactants can be found by considering the acoustic wave equation with chemical source term. Neglecting the Doppler effect and the gradients in the mean flow (Clavin and Searby (2016)):

$$\frac{\partial^2 p}{\partial t^2} - a^2 \Delta p = \frac{\partial \dot{q}_{\gamma}}{\partial t} \tag{8.1}$$

where $\dot{q}_{\gamma} = (\gamma - 1)\dot{q}_v$ and \dot{q}_v is the rate of heat release per unit volume. Eq. (8.1) shows that if a pressure wave forms inside the hot spot, it can gain in amplitude if a source of \dot{q}_{γ} (a reaction wave for instance) can follow the pressure wave at the same speed (speed of sound). This situation corresponds to a thermo-acoustic like instability, and can be achieved by a gradient of induction time. Indeed, consider a hot kernel of reactants of radius r_0 preconditioned by a gradient of τ_I (see Fig. 8.2). Contrary to the homogeneous case, a reaction wave forms at the point of minimum τ_I and propagates towards larger τ_I at speed u_I , which is directly linked to the gradient of induction time via $u_I = (d\tau_I/dx)^{-1}$. This induction front induces an instantaneous distribution of \dot{q}_v , thereby generating a perturbation of the pressure field. A compression wave thus forms, which propagates at the speed of sound a. Similarly to the Rayleigh criterion, an instability is triggered if both the acoustic wave and the induction front are synchronize, i.e. if u_I is close to a. As soon as the pressure wave exits the hot spot, the term \dot{q}_{γ} vanishes and the pressure build-up stops. Therefore, the critical value $P_{N_{CJ}}/P_u$ may be reached only if chemical heat can be released on a time scale τ_E lower than the residence time τ_a of the pressure wave inside the hot spot. τ_a is defined by $\tau_{\mathbf{a}} = r_0/a$, where r_0 is the radius of the hot spot.

The evolution of the hot spot is then totally determined by the four variables $(\mathbf{u}_{\mathbf{I}}, \mathbf{a}, \tau_{\mathbf{E}}, \tau_{\mathbf{a}})$. What is important here is not the parameters themselves but how they compare with each other. Therefore, the size of the problem can be further reduced to two dimensions: $(\varepsilon, \xi) = (\tau_a/\tau_E, a/u_I)$. ξ accounts for the resonance between the acoustic wave and the reaction front. ε measures the chemical energy fed to the acoustic wave during its propagation inside the hot kernel eventually leading to a rapid increase of pressure. Bradley and collaborators (Bradley and Kalghatgi (2009); Bates et al. (2016); Bates and



Figure 8.3: Outcome of a hot spot described in the plane (ε, ξ) by Bradley and collaborators (Bradley and Kalghatgi (2009); Bates et al. (2016); Bates and Bradley (2017)).

Bradley (2017)) used the (ε, ξ) plane to characterize the evolution of the hot spot. As illustrated in Fig. 8.3, the plane can be divided into a number of regions:

- For $\varepsilon < 1$: Any deposition of chemical energy occurs on a time scale higher than the acoustic time and no local runaway is possible;
- For ε ≥ 1: Chemical energy is deposited on a time scale lower than the acoustic time and a rapid increase of pressure can be observed provided a synchronization of the acoustic wave and the reaction front is obtained. This coupling depends on the value of ξ:
 - For $\xi < \xi_l$: the induction time gradient is too low and the system reduces to the homogeneous constant volume kernel. Thus the pressure increase is too low to create a CJ wave as explained earlier;
 - For $\xi > \xi_u$: the induction time gradient is too steep and the system reduces to a constant pressure combustion.
 - For $\xi_{l} \leq \xi \leq \xi_{u}$: the spontaneous initiation of a detonation can be observed as shown by Bradley and Kalghatgi (2009). For low values of ε , the amount of chemical energy fed to the pressure wave, during its propagation in the hot spot, is small so that a quasi perfect resonance between acoustics and heat release is necessary, hence the narrow tip of the peninsula. This region widens for larger values of ε because of the strong chemical energy input, thereby allowing less restrictive conditions on the coupling.

8.2 On the difficult process of generating hot spots

8.2.1 Taking into account the deflagration time scale

Section 8.1 showed that a detonation can emerge from a preconditioned hot spot, provided that acoustic and induction time scales are comparable. However, the explosions discussed in this thesis involve the propagation of very fast flames, with very small associated time scales. Therefore, if one wants to explain DDT with the Zeldovich mechanism, the time scale of the deflagration τ_f must be taken into account. We show, in the following, that



Figure 8.4: Temporal evolution of the maximum temperature of the reactants in the case $\phi = 0.52$ presented in Chapter 7.

bringing the mixture to an ignitable state is not an easy task in the case of fast flames. Any point in the unburnt side, heated to a temperature T_u^* , can be ignited if the induction time $\tau_I(T_u^*)$ is lower than the time necessary for the flame to reach that particular point and burn the reactants. τ_I is typically a decreasing function of T_u^* , so that there exists, for highly reactive mixtures, a minimum temperature $[T_u^*]_{min}$ above which τ_I is lower than τ_f and ignition of the mixture is possible. Let us apply this discussion to the case $\phi = 0.52$ discussed in Chapter 7, to understand why no thermal explosion occurred. Shocks formed in the obstructed region, as highlighted in Section 7.2.2.2, leading to a stepwise increase of the fresh gases temperature. The temporal evolution of the maximum temperature of the reactants $T_{u,max}$ is displayed in Fig. 8.4. It shows that the reactants are indeed heated by the shock train going from the initial 300K to the maximum value $\approx 520K$. This temperature obviously corresponds to a frozen state.

Let us now give an order of magnitude of the temperature T_u^{\star} that would be necessary in the case $\phi = 0.52$ to observe ignition. First, a definition is needed for τ_f that accounts for the acceleration of the flame:

$$\tau_f = \mathcal{L}/s_{tip}$$

where \mathcal{L} is some characteristic length scale of the problem, which in this case can be taken as the space separating two consecutive obstacles: $\mathcal{L} = \mathcal{S}$. The evolution of τ_f with the flame position is displayed in Fig. 8.5. During the flame propagation in the obstructed region, τ_f decreases by two orders of magnitude due to the exponential acceleration of the flame. At the end of the obstructed region, τ_f is very close to typical induction times at high temperatures, say higher than 1000 K, which confirms the necessity to include τ_f when discussing DDT in the case of fast flames. τ_f is used to evaluate the minimum temperature $[T_u^{\star}]_{min}$, above which the condition $\tau_I \leq \tau_f$ is reached for two pressure conditions: P = 1 bar corresponds to the pressure condition before ignition of the mixture and P = 3 bar is slightly higher than the maximum mean pressure inside the chamber during the flame propagation in the obstructed region. As soon as the flame reaches the middle of the obstructed zone ($\approx 1 \ m$), $[T_u^{\star}]_{min}$ becomes higher than 950 K for both pressure conditions. The large gap between $[T_u^*]_{min}$ and $T_{u,max}$ ($\approx 400 \text{ K}$) demonstrates that DDT via the Zeldovich mechanism is not possible in this case. This shows that, for fast deflagrations, forming hot kernels of reactants is not an easy task and requires much stronger shocks than the ones observed in the case $\phi = 0.52$.


Figure 8.5: Evolution of the flame propagation time scale $\tau_f = S/s_{tip}$ as a function of the flame tip position X_{tip} in the obstructed region. The minimum fresh gas temperature $[T_u^*]_{min}$, above which the induction time becomes lower than τ_f , is also displayed for two pressure conditions (1 bar and 3 bar).

8.2.2 The capacity of faster flames to generate hot spots

Heating the combustible to a temperature higher than $\approx 950 \ K$ appears to be a necessary condition to ignite the mixture. This requires much stronger shocks than the ones observed in the case $\phi = 0.52$. If the flame brush can be brought to a sufficiently higher propagation speed, such discontinuities can be generated. To prove this, let us consider an isobaric compression wave formed ahead of the moving piston. Its temperature can be evaluated from the relation (Clavin and Searby (2016)):

$$(T/T_u)^{1/2} = 1 + (\gamma - 1)(u/a_u)/2$$
(8.2)

where u is the flow velocity ahead of the deflagration and is the only variable of the problem. Increasing u to a sufficiently high value, much greater that the sound speed a_u , can increase the temperature of the reactants to values greater than 550K. Larger u, compared to the case $\phi = 0.52$, can be obtained by considering mixtures with higher reactivity, thereby reaching faster deflagrations. This conclusion is consistent with the experimental results. Figure 8.6(left) displays the evolution of the flame propagation speed s_{tip} with the flame position X_{tip} for two equivalence ratios $\phi = 0.52$ and 1. It shows the drastic increase of s_{tip} with increasing ϕ , which results in DDT just prior to the fifth obstacle for $\phi = 1$. Figure 8.6(right) shows that the part of the s_{tip} curves corresponding to FA (i.e. before the DDT point in the stoichiometric case) collapse when normalized by the laminar flame speed s_L , which indicates that s_L is the dominant parameter of the FA process. Therefore, richer mixtures can lead to higher flame propagation speeds, hence higher flow velocity ahead of the deflagration.

Let us now confirm, using the experimental data, that stronger shocks are indeed produced in richer mixtures because of the higher flow velocity. Figure 8.7 displays the



Figure 8.6: Influence of the mixture equivalence ratio ϕ on the explosion scenario (experimental results). $\phi = 0.52$ and $\phi = 1$ correspond to a deflagrating and a detonating case respectively. In the right figure, the flame propagation speed s_{tip} is normalized by the laminar flame speed to show that s_L is the dominant parameter of the flame acceleration process here.

overpressure record for probe 2 located very close to the DDT locus (its position is indicated in Figure 8.6(left)). For $\phi = 1$, the probe captures the emerging detonation front as well as the strong shocks prior to DDT. In both experimental shots provided by the Gravent database, an Incident Shock (IS $\stackrel{\sim}{\searrow}$) appears a few microseconds before the detonation is detected with an overpressure peak (4.3bar) approximately 2 times larger than the pressure peaks detected by the same probe in the deflagrating case (see Fig. 7.4). IS reflects off the fifth obstacle, placed just 50 mm downstream of the probe, and later reappears (*) with a much higher pressure peak around 13 bar.

As discussed in Section 6.2, the range of equivalence ratios considered in the Gravent explosion channel corresponds to the region of increasing flame consumption speed s_L . DDT is systematically observed for equivalence ratios higher than a certain threshold value. This leads to the following conclusions: 1) reaching a supersonic regime of propagation is not a sufficient condition to trigger DDT; 2) there exists a critical value of the piston speed, hence flow velocity, above which sufficiently high compression can be achieved, which is consistent with Eq. (8.2); 3) Strong shocks, and more importantly their reflection at the walls, are a key ingredient for a successful detonation initiation in obstructed channels.

8.3 The capacity of strong shocks to trigger DDT

As discussed above, strong shocks are of crucial importance to the DDT process in obstructed channels. We still need to identify the mechanisms by which such discontinuities can trigger DDT.



Figure 8.7: Overpressure signal at probe 2 located 1.4 m away from the ignition plate. (Left) Influence of the equivalence ratio of the pressure buildup. (Right) Zoom on the instants surrounding the arrival of the detonation front for two experimental shots at $\phi = 1$.

8.3.1 Thermal explosion

As discussed in Section 8.1, a detonation front can emerge from a hot kernel provided some preconditioning of the reactive spot is obtained. If the aforementioned shocks are strong enough, they can bring the reactants to sufficiently high temperatures, typically higher than 1000 K for hydrogen/air mixtures. It is important to note that achieving "high" temperatures is of crucial importance for a successful detonation initiation, since He and Clavin (1992) identified a quenching mechanism at relatively "lower" temperatures. What remains to be identified is the process that results in the formation of a spot of induction time gradient.

Thanks to a number of very accurate numerical simulations, Oran and coworkers have shown the capacity of shock reflections to produce such a preconditioning. They are summarized in the review paper of Oran and Gamezo (2007). The interaction of a Reflected Shock (RS) with the flow induced by the Incident Shock (IS) is illustrated in Fig. 8.8. The wall can typically represent one obstacle in the BR30hS300 channel. Because of viscous heating, IS leaves behind a slightly heated and growing in time boundary layer. When IS reflects off the wall, it interacts back with the boundary layer, forming a bifurcated shock structure composed of: 1) the main reflected shock; 2) an oblique leading shock (or bifurcated foot); 3) a tail shock downstream. The bifurcated shock grows in time as it expands in the transverse direction. The temperature distribution behind this structure can be strongly nonhomogeneous. In the numerous numerical simulations of DDT summarized in Oran and Gamezo (2007), thermal explosions have been observed mainly in two possible locations, illustrated in Fig. 8.9. In the top frames, DDT occurs at the bottom wall where the tail shock interacts with the bottom boundary. In the bottom frames, the temperature behind the main reflected shock is higher than in the remaining part of the channel. Furthermore, the temperature increases with distance from the shock. This temperature gradient results in the spontaneous initiation of a detonation from the point of highest temperature, i.e. the point of lowest induction time.



Figure 8.8: An incident shock (left) reflects off the wall and interacts back with the flow it induced. The wall here can typically represent an obstacle in the BR30hS300 channel. From Oran and Gamezo (2007).

8.3.2 Flame/shock interaction

Strong shocks can also induce DDT via another mechanism revealed recently by Goodwin et al. (2016a,b) using numerical simulations. It does not require to bring the mixture to an ignitable state. It is illustrated in Fig. 8.10. An incident shock, just ahead of the flame brush, reflects of the bottom wall to interact back with the combustion wave. A few microseconds later, a detonation emerges from the inner structure of the flame. Therefore, the flame plays a crucial role in this mechanism. It is worth noting that, even though the scenario is different from the spontaneous initiation of a hot pocket of fresh mixture, the Zeldovich mechanism can not be fully ruled out since a temperature gradient is provided by the flame structure. This mechanism will be discussed in more details in Chapter 9.

8.3.3 Runaway mechanism

Another mechanism worth mentioning was first suggested by the theoretical study of Deshaies and Joulin (1989). Consider the simple one-dimensional flame/shock complex: a weak shock propagating at constant velocity D followed by a flame propagating at constant speed U_f . Since U_f is influenced by the temperature of the fresh mixture perturbed by the leading shock, a positive feedback can take place leading to the increase of the flame speed. Deshaies and Joulin (1989) showed that there exits a critical flame speed U_f^* above which the flame can not sustain the high propagation regime and a thermal runaway of the flame speeds occurs, leading to DDT. To bring the flame speed to the value U_f^* , which is typically supersonic, flame acceleration mechanisms are required and can be any of those identified in Chapter 7 for example.

Recent experiments in smooth channels, performed by Kuznetsov et al. (2010), revived interest in this process. These experiments considered highly reactive mixtures, namely stoichiometric hydrogen/oxygen and ethylene/oxygen flames, initially at 293 K and 0.5 – 0.75 bar. Prior to DDT, the flame propagated behind a weak shock at a velocity $U_f \approx$ 500 m/s. The temperature of the reactants trapped between the deflagration and the leading shock was too low to induce ignition \approx 550 K. However, even in these conditions, DDT was observed inside the flame brush.

These experiments motivated Kagan and Sivashinsky (2017) to revisit the Deshaies and Joulin (1989) model. They introduced a cut-off temperature $T^* = 750 \ K$ so that no



Figure 8.9: Two possible locations for the spontaneous initiation of a detonation behind a bifurcated shock structure. (Top) Instantaneous density, temperature and pressure fields that shows DDT occurring at the bottom wall where the tail shock interacts with the boundary layer. (Bottom) Temperature fields at different instants surrounding DDT showing the formation of a gradient of temperature behind the reflected shock and the thermal explosion starting at the right wall where the temperature is the highest. From Oran and Gamezo (2007).



Figure 8.10: Temperature fields for different instants surrounding DDT. The latter results from the interaction of a reflected shock with the flame brush. Time in milliseconds is shown in the frame corner. From Goodwin et al. (2016b).

chemical heat could be released for $T < T^*$. The temperature of the unburned material trapped between the flame and the leading shock never exceeded T^* . These gases are therefore in a frozen state, similarly to the Kuznetsov et al. (2010) experiments. A transition to detonation was still observed and can not be the result of a spontaneous initiation. Instead, DDT resulted from a thermal runaway of the flame speed.

8.4 No universal mechanism

As discussed above, there are a number of different mechanisms that can explain the complex DDT problem. It is true that the Zeldovich mechanism, hence the thermal explosion, is a sufficient condition to trigger a detonation. However, one may argue that it is not a necessary one. Moreover, meeting the requirements for the spontaneous initiation of hot spots is a delicate compromise, which requires the appropriate reactivity gradient

as well as high temperatures to avoid some quenching mechanisms at lower temperatures (see He and Clavin (1992)). Finally, there is no reason to believe that the DDT mechanism is either unique or universal. DDT in complex configurations is an intricate process that may involve a combination of different mechanisms, the interplay among which can lead to less restrictive conditions for the successful onset of detonations.

Chapter 9

Numerical investigation of the DDT mechanisms in the stoichiometric case

Abstract

In Chapter 6, two hydrogen/air explosion scenarios were selected from the BR30hS300 database: a very lean mixture ($\phi = 0.52$) where only flame acceleration was observed and a stoichiometric case where DDT was triggered. The present chapter presents a numerical investigation of the detonating case. More specifically, it focuses on the impact of the chemistry modeling on the DDT scenario. This study is motivated by recent numerical investigations of DDT using detailed chemistries Ivanov et al. (2011), which suggested that the mechanism responsible for the transition may be different from the one usually observed using single-step reaction models. Both approaches to chemistry modeling have been selected in this study to perform the simulation. The numerical investigation proceeds in two steps. The first part focuses on DDT and the results are presented in the form of an article submitted to the international journal Combustion and Flame. The second part is dedicated to the dynamics of the self-propagating detonation. Large discrepancies are observed between the detonation structure obtained with the detailed chemistry and the one observed using the single-step reaction model. The chapter ends with an attempt to explain these differences.

9.1 Focus on DDT

In this chapter, two changes in notations are introduced that are dictated by the submitted article. First, Y corresponds now to the vertical direction. Second, the progress variable c is now given by $c = Y_{H_2}/Y_{H_2,u}$, such that it is equal to 1 in the fresh gases and 0 in the burnt gases. The authors apologize for this sudden change in notations.

Influence of kinetics on DDT simulations

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Abstract

Deflagration to Detonation Transition (DDT) is an intricate problem that has been tackled numerically, until recently, using single-step chemical schemes. These studies [1] showed that DDT is triggered when a gradient of reactivity forms inside a pocket of unreacted material. However, recent numerical simulations of hydrogen/air explosions using detailed reaction mechanisms [2] showed that detonation waves can emerge from the flame brush, unlike what was usually seen in the single-step simulations. The present work focuses on chemistry modeling and its impact on DDT. Using the idealized Hot Spot (HS) problem with constant temperature gradient, this study shows that, in the case of hydrogen/air mixtures, the multi-step chemical description is far more restrictive than the single-step model when it comes to the necessary conditions for a hot spot to lead to detonation. A gas explosion scenario in a confined and obstructed channel filled with an hydrogen/air mixture is then considered. In accordance with the HS analysis, the Zeldovich mechanism [3] is responsible for the detonation initiation in the single-step case, whereas another process, directly involving the deflagration front, initiated DDT in the complex chemistry case. In the latter, the detonation wave starts in the flame brush through Pressure Pulse (PP) amplification. It is shown that the presence of highly reactive species in the detailed chemical schemes is responsible for the formation of the pressure pulse that initiates DDT.

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1. Introduction

Mining, process and energy industries suffer from billions of dollars of worldwide losses every year due to gas explosions. In addition to these costs, explosion accidents are often tragic and lead to severe injuries and fatalities. These dam-

ages vary significantly from an accident to another, depending on the explosion scenario. A sound knowledge of explosion physics is therefore of vital importance for the prediction of hazard potentials and the implementation of efficient preventive measures.

Gas explosions have been studied experimentally [1, 4], theoretically [3, 5–10]

- ¹⁰ and numerically [1, 11–13]. They occur when a reactive mixture is subjected to an external source of energy from which a combustion wave emerges. This reaction wave can have two possible propagation modes [14, 15]: supersonic in the case of detonation and subsonic in the case of deflagration. The direct initiation of detonation has been studied experimentally and theoretically [16–19] and is
- only possible if a sufficient energy is deposited quasi-instantaneously. In most cases, this condition is not met and a deflagration emerges from the ignition source.

Deflagrations are intrinsically unstable and tend to accelerate continuously after ignition [15]. This Flame Acceleration (FA) process is well understood thanks to

- extensive experimental (summarized in the Ciccarelli and Dorofeev review paper [4]), theoretical [5–7] and numerical [11, 12] works and depends on the degree of confinement and obstruction encountered by the flame during its propagation. FA can either result in fast deflagrations or create the appropriate conditions for the Deflagration-to-Detonation Transition (DDT). Understanding DDT is
- ²⁵ crucial because it often leads to an escalation in the hazard scenario. Years of theoretical and numerical efforts, summarized in [1], have been devoted to exploring the different mechanisms that can initiate DDT. Despite this, a first

principle description of the relative importance of these mechanisms, and the possible interplay among them, is still incomplete.

- ³⁰ During flame acceleration, the reaction front acts like a piston [14] moving the reactants ahead of it. As a result of the displacement flow, compression waves/shocks are formed ahead of the flame. Thus, a propagating deflagration wave usually consists of the reaction front following a precursor compression wave/shock. This deflagration-shock complex has been tackled by many inves-
- tigators [8, 9, 15]. They showed that: 1) the precursor shock is, in general, faster that the deflagration; 2) once the deflagration reaches a critical speed, equal to the sound speed of the burned gases, there exists a path along which a deflagration can reach a velocity equal to that of the precursor shock, forming a self-sustained detonation wave (or CJ detonation). This path is referred to as
- ⁴⁰ the generalized Hugoniot curve by Troshin [10] and basically describes all the successive solutions for the double discontinuity problem during DDT. In other words, according to this simple approach to DDT, the deflagration can simply accelerate to merge with the leading shock wave and form a detonation. A breakthrough in detonation research occurred when Oppenheim and Urtiew
- ⁴⁵ [8, 20], later backed up by the simulations of Oran and coworkers [1], showed that the detonation onset is not simply the result of the coalescence between a flame and a shock. Instead it was observed that the reactants trapped between the leading shock and the deflagration are heated and compressed to the point where enough energy is accumulated and an explosion can occur. These
- ⁵⁰ observations are consistent with the mechanism proposed by Zeldovich *et al.* [3] and later observed experimentally by Lee *et al.* [21]. Consider a reactive mixture exposed to a gradient of induction time $\nabla \tau_I$, which may be a gradient of temperature or pressure or reactants concentration. A reaction wave emerges from the point of lowest induction time τ_I and its propagation speed depends on
- ⁵⁵ $\nabla \tau_I$. For a steep initial gradient, a weak shock forms and quickly detaches from the reaction wave which proceeds as a normal flame. Conversely, for a small initial $\nabla \tau_I$, the mixture is simply uniformly ignited without shock formation. For intermediate values of $\nabla \tau_I$, the reaction wave and the compression waves

can propagate in phase. In this case, the coherent energy release causes the compression wave to steepen into a shock. The amplification of the latter and

its coupling to a combustion wave later forms a detonation.The presence of obstacles in the flame path is known [1, 22] to facilitate DDT.First, obstructions enhance FA thus producing strong shocks ahead of the flame.Second, these shocks can interact with the obstacles allowing shock focusing at

the corners, shock reflections and collisions and shock flame interactions. These events create the appropriate conditions for the detonation onset. This has been the major trend in DDT research until a recent paper of Kuznetsov et al. [23]. Evaluating the Mach of the shock ahead of the flame prior to DDT, they showed that the temperature of the preheat zone ahead of the flame (550K)

- for a stoichiometric hydrogen/oxygen mixture) does not allow spontaneous ignition. Therefore, the Zeldovich mechanism could not explain DDT in this case. Revisiting the Deshaies-Joulin [24] analysis and taking into account foldinginduced flame acceleration, Kagan and Sivashinsky [25] showed that a positive feedback between the flame and the flame-driven precursor shock can explain
- ⁷⁵ DDT in Kuznetsov *et al.* experiments. The flame, unable to sustain high displacement speeds, simply transitions into a supersonic propagation mode (detonation). Finally, Ivanov *et al.* [2] showed that the scenario leading to DDT, when using detailed chemistry modeling, may differ from the classical Zeldovich mechanism observed using single-step chemistry, which is the classical approach
- ⁸⁰ to DDT simulations.

These last findings suggest that the Zeldovich mechanism is not a universal trigger for DDT, and that the scenario leading to DDT may depend greatly on the chemistry modeling used in numerical simulations. This motivated the present study, which focused on the influence of kinetics on DDT using two-

dimensional DNS. The configuration considered is based on the Gravent explosion channel database [26]. The confined and obstructed channel, presented in Section 2, is filled with a quiescent stoichiometric hydrogen/air mixture. The 9S18R (9 species and 18 irreversible reactions) and 3S1R (3 species and 1 reaction) chemical schemes used in this paper are presented in Sections 3.1 and



Figure 1: Sketch of the Gravent explosion channel geometry [26]. A series of 7 obstacles are evenly placed at the top and bottom plates of the channel. The constant spacing S is equal to 300 mm and the blockage ratio is Br = 2h/H = 30%. From the last obstacle to the end plate, the channel is unobstructed allowing detonation propagation without perturbation. Distances are in millimeters.

3.2. These mechanisms are first compared using the canonic Hot Spot (HS) case with constant temperature gradient in Section 3.4. This allows to investigate the requirements, in terms of temperature and radius, for HS to trigger DDT using 3S1R and 9S18R. The conclusions will be used to explain the results of the simulations of the BR30hS300 channel, presented in Section 4. The impact of the chemistry modeling on the DDT mechanism is analyzed in Section 4.3.

2. Numerical setup

The 2D direct numerical simulations presented in this paper are performed using the DNS/LES solver for the fully compressible multispecies Navier-Stokes equations AVBP, co-developped by CERFACS and IFPEN [27]. A centered continuous Taylor-Galerkin scheme third-order in space and time (TTGC [28]) is used.

To investigate DDT in obstructed and confined channels, the BR30hS300configuration of the Gravent database [26] is chosen (Fig. 1). The channel is $H = 60 \ mm$ high and $L = 5400 \ mm$ long. 7 obstacles are evenly spaced along the top and bottom walls of the channel with a constant blockage ratio Br = 2h/H = 30%, where h and H are the height of an obstacle and the channel respectively. The spacing S between the obstacles axis is equal to 300 mm.



Figure 2: Illustration of the interpolation procedure between the different meshes used for the present simulations. The interpolation from mesh to mesh is performed when the flame is separated by a distance of two spacings 2S from the end of the fine section of the mesh. $t_{Interp}(1 \rightarrow 2)$ denotes the time when the interpolation, from the first mesh to the second, is performed.

From the last obstacle (X = 2.05m) to the closed end wall (X = 5.4m), the section of the channel is constant allowing detonation propagation without per-

- turbation. A three-dimensional DNS of a channel this large is not affordable. Only a two-dimensional longitudinal slice of the BR30hS300 configuration is therefore considered. Note that 2D simulations of DDT are still largely used nowadays [1] to explore the mechanisms involved in DDT. All obstacles and walls are treated as no-slip adiabatic boundaries.
- ¹¹⁵ Usually an Adaptive Mesh Refinement (AMR) is used when simulating DDT in large channels. While this strategy is useful to maintain reasonable computational costs, it requires an a priori knowledge on the key ingredients impacting the problem, thereby increasing the resolution in the regions where these mechanisms activate. Here, another strategy, based on multiple meshes, is used (see
- Fig. 2). Each grid is stepwise homogeneous and composed of two zones: 1) a refined section, 4.5S large, where the resolution is uniform $\Delta x_{fine} = 20 \ \mu m$, thereby allowing to capture the deflagration front (350 450 μm thick), the detonation front (half reaction thickness: 153 μm) as well as the flow structures;

2) a coarser grid, starting at least $2S = 600 \ mm$ ahead of the combustion wave, ¹²⁵ with a uniform and larger element size $\Delta x_{coarse} = 120 \ \mu m$. When the distance separating the tip of the combustion wave from the end of the refined section reaches 2S (Fig. 2(b)), a piecewise linear interpolation of the solution is performed to another mesh with the same characteristics as the previous one except that the refined section now starts just behind the flame tail (Fig. 2(c)).

- ¹³⁰ This interpolation is performed 3 times during the flame propagation in the obstructed region without any noticeable numerical noise. It allows to correctly capture the combustion waves as well as the flow structures far ahead of the flame.
- The channel is filled with a quiescent mixture of hydrogen and air at atmospheric conditions. It is ignited at the left closed end using a semi-spherical 5 mm radius hot kernel. This initialization procedure was employed in other gas explosion studies ranging from venting deflagrations [11, 13] to DDT [1, 12]. Among all the equivalence ratios considered in the experiments, the case corresponding to stoichiometric hydrogen/air and showing DDT was selected.

¹⁴⁰ 3. Chemistry modeling

The objective of the present paper is to study the influence of the kinetics modeling on DDT. To do so, a single-step reduced chemistry, which is the classical approach for DDT simulations, is confronted to a multi-step mechanism. Besides the chemical scheme, the numerical setup is the same for all simulations.

145 3.1. Detailed chemistry

The multi-step mechanism chosen to model hydrogen/air chemistry is the mechanism derived by Boivin *et al.* [29], composed of 9 species and 18 irreversible reactions. This mechanism is referred to as 9S18R in the following. 9S18R is validated against experimental data for atmospheric hydrogen/air lam-

inar flame speeds s_L for a wide range of equivalence ratios $\phi \in [0.4..1.5]$ in Fig. 3(left). The mechanism predicts s_L with very good accuracy at stoichiome-



Figure 3: Left: One dimensional hydrogen/air laminar flame speed s_L at atmospheric conditions for various equivalence ratios ϕ . (Solid line) 9S18R mechanism [29]; experimental data: (\Box) Berman [30], (\circ) Dowdy *et al.* [31], (\bigtriangledown) Egolfopoulos *et al.* [32], (\triangle) Iijima *et al.* [33], (+) Takahashi *et al.* [34]. Right: Influence of temperature and chemistry on the induction time τ_I and the excitation time τ_E for $\phi = 1$ and $P = 1atm. \tau_I$ from experiments: (\Box) Slack *et al.* [35] and (\circ) Snyder *et al.* [36]. T_{9S}^c and T_{3S}^c are the ignition threshold temperatures for the 9S18R and 3S1R mechanisms respectively.

try, which corresponds to the case considered for the DDT simulations presented in Section 4.

3.2. One-step reduced chemistry

ulations presented in Section 4.

¹⁵⁵ Until very recently, the most common approach in DDT simulations has been the use of reduced single-step mechanisms [1]. The methodology to derive such chemistries using only one transported species is detailed in [37]. The same procedure was later used by Wang *et al.* [38] to derive a one-reaction mechanism for hydrogen/air flames, taking into account the reaction order. This strategy was selected to develop the single-step chemistry, named 3S1R, used in the sim-

3S1R is designed to reproduce both the flame and the detonation global features. Two fictive species S_1 and S_2 are used, and the reaction formula reads $S_1 + 0.5S_2 \rightarrow \mathcal{P}$, where \mathcal{P} denotes the products. This allows to mimic the

- hydrogen oxidation reaction. S_1 and S_2 have the same molecular weight Wequal to the average molecular weight of an hydrogen/air mixture at stoichiometry, i.e. W = 0.021 kg/mol. Therefore W is constant across the flame. The reactants share the same specific heat capacities at constant pressure C_p and constant volume C_v so that the specific heat ratio γ is constant across the flame front. The Lemis number of the specific Lemma is set to unity. The mean
- flame front. The Lewis number of the species Le_{S_k} is set to unity. The reaction rate of the irreversible reaction is modeled using an Arrhenius formulation: $\dot{\omega} = A(\rho/W)^{1.5}Y_{S_1}(Y_{S_2})^{0.5}exp(-T_a/T)$. The set of target parameters are:
 - The laminar flame speed s_L and the adiabatic flame temperature T_b : they allow the reduced mechanism to reproduce the effect of reactants consumption and gas expansion on the flame propagation during the FA phase. s_L and T_b are imposed equal to the one obtained with the 9S18R mechanism at $\phi = 1$.
 - The Chapman-Jouguet detonation speed D_{CJ} and the half detonation thickness δ_d given by the ZND model for the detonation structure. δ_d can be linked to the detonation cell size via empirical relations [1].
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To match these targets, a set of parameters can be tuned: the specific heat ratio γ , the chemical energy release Q, the pre-exponential factor A and activation temperature T_a . The values $\gamma = 1.17$ and $Q = 43.28 R T_0/M J/kg$ result in the desired targets: $T_b = 7.13T_0$ and $D_{CJ} = 1993m/s$, $T_0 = 300K$ being the fresh gas temperature and M the mean molar mass of the mixture. Computing the 1D laminar flame in AVBP and the theoretical ZND model using the selected values of γ and Q, the chemical parameters A and T_a are determined so that $s_L = 2.43m/s$ and $\delta_d = 153\mu m$. The reaction rate model then reads $\dot{\omega} = 3.9 \times 10^7 \ (\rho/W)^{1.5} Y_{S_1} (Y_{S_2})^{0.5} exp(-46.36T_0/T) \ kg/m^3/s$.

¹⁹⁰ 3.3. Influence of chemistry on ignition characteristic times

DDT involves spontaneous ignition. Therefore, the influence of the chemical scheme on the induction time τ_I and the excitation time τ_E has to be investigated. τ_I is defined as the time for which an homogeneous mixture must be maintained at a given temperature and pressure before exothermic reactions activate. After τ_I , the heat release rapidly blows up and τ_E is defined as the 195 time necessary for the heat release to go from 5% to 100% of its peak value. τ_I and τ_E obtained with 9S18R and 3S1R are displayed in Fig. 3(right) for temperatures in the range [600..1400]K and atmospheric pressure ($\phi = 1$). The τ_I dependance on temperature obtained with the 9S18R mechanism is in good agreement with the experimental data. The ignition threshold T^c is defined 200 here as the temperature below which the induction time needed before ignition is higher than the longest time scale of flame propagation in the BR30S300 configuration $\tau_f = L/s_L \approx 2.2ms$. This means that when the temperature of the reactants is below T^c , any point on the fresh mixture can be burned by the flame before it can ignite. The ignition threshold $T_{9S}^c \approx 950 K$ obtained 205 with the nine-step scheme agrees reasonably well with the experimental data. Conversely, the induction times obtained using the reduced mechanism are orders of magnitude smaller than the experimental data, which coincides with the conclusions of Ivanov *et al.* [2] regarding τ_I obtained with reduced chemistries

for stoichiometric H_2/O_2 mixture. Note that, with 3S1R, the mixture can be



Figure 4: Left: A sketch of the hot spot (HS) of radius r^0 with constant temperature gradient. The reaction wave emerges from the point of minimum ignition time τ_I . Right: The $\xi - \varepsilon$ diagram shows the detonation peninsula [39]: DDT is possible between the upper branch ξ_u and the lower branch ξ_l .

ignited even for low temperatures, about 300 K lower than T_{9S}^c . Like τ_I , τ_E obtained with 3S1R are much lower than the ones obtained with 9S18R. The impact of these findings on DDT will be discussed in Section 3.4.

3.4. Influence of the chemistry modeling on a canonic DDT problem

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In this section, a comparison between the two chemical models is performed with regard to DDT caused by the Zeldovich mechanism [3]. The latter has been designated by a series of DNS [1] as a sufficient condition for DDT onset. The canonic HS problem, sketched in Fig. 4(left) is considered in the following.

220 3.4.1. The hot spot problem and the detonation peninsula

According to Zeldovich and co-workers, a detonation can emerge from a hot spot, i.e. a region characterized by a gradient of induction time $\nabla \tau_I$ [3]. Inside the hot spot, a combustion wave forms at the point of minimum τ_I . If $\nabla \tau_I$ is too steep, a shock forms and separates from the combustion wave. The latter

proceeds as a deflagration. On the other hand, if $\nabla \tau_I$ is too small, the entire hot spot is ignited uniformly. For an intermediate range of $\nabla \tau_I$ however, a coupling between the shock and the combustion wave is possible. A positive feedback between the two waves eventually leads to the formation of a detonation front. Bradley *et al.* [39] characterized the gradient of induction time $\nabla \tau_I$ needed to

- trigger DDT. Inside the hot spot, both a reaction wave and a compression wave are formed. To allow a resonance between both waves, the reaction wave velocity $u_I = dx/d\tau_I$ must neither be too high nor too low compared to the compression wave velocity (sound speed) a. A comparison between u_I and a can be made through the variable $\xi = a/u_I$. For certain values of ξ allowing resonance, the
- compression wave can gain in amplitude if the heat release inside the reaction wave is deposited fast enough. This can be characterized by introducing the variable $\varepsilon = r^0/(a\tau_E)$, where ε measures the chemical energy fed to the pressure pulse during its residence inside a hot spot of radius r^0 . A region in the $\xi - \varepsilon$ space, called the detonation peninsula [39], has been identified as the region
- where the conditions at the hot spot are ideal for the onset of detonation. The region is illustrated in Fig. 4(right), between the upper branch ξ_u and the lower branch ξ_l .

3.4.2. The influence of chemistry modeling on the hot spot scenario

- In this section, the $\xi \varepsilon$ diagram is used to investigate the outcome of a hot spot using the 9S18R and the 3S1R mechanisms. Consider a hot spherical kernel with a constant temperature gradient and a non-dimensional size $h^* = 2r^0/h$, where h is the height of an obstacle in the BR30hS300 channel (see Fig. 4). The non-dimensional peak temperature inside HS is defined as $T^* = (T^0 - T^c)/T^c$, where T^c is the ignition threshold. Negative values of T^* indicate temperatures
 - at which the mixture will not be ignited. Since the values T^c obtained with 9S18R agree very well with the experimental data, T^c is set equal to T_{9S}^c .

The $\xi - \varepsilon$ plane offers two limit lines: 1) the minimum energy input $\varepsilon_c = 1$, 2) the perfect resonance between compression and combustion waves $\xi_c = 1$.

These limits allow to define critical hot spot parameters that can trigger DDT: 1) h_c^{\star} is the minimum non-dimensional HS size where pressure waves can travel



Figure 5: The influence of pressure P and temperature T^* on critical HS parameters for the single-step (dashed lines) and the multi-step mechanisms (solid lines). Left: h_c^* is the minimum non-dimensional size that allows energy input to pressure waves, i.e. $\varepsilon_c = 1$. Right: $[\partial T/\partial r]_c^*$ is the minimum non-dimensional temperature gradient that allows resonance between pressure and reaction waves, i.e. $\xi_c = 1$.

to gain energy, i.e. $h_c^{\star} = 2\varepsilon_c(a\tau_E)/h$; 2) ξ can be reformulated [39] as $\xi = a(\partial T/\partial r)(\partial \tau_I/\partial T)$ in the radial coordinates, thereby the minimum HS temperature gradient that leads to a resonance between pressure and reaction waves can

- be expressed as $[\partial T/\partial r]_c = \xi_c/(a\partial \tau_I/\partial T)$. A non dimensional form for $[\partial T/\partial r]_c$ is then $[\partial T/\partial r]_c^* = (T^0/h_c)[\partial T/\partial r]_c$, where $h_c = h_c^*h$. h_c^* and $[\partial T/\partial r]_c^*$ depend on the conditions of pressure P and temperature T^* inside the hot spot. The influence of P and T^* as well as the chemical scheme on the critical HS parameters is assessed in Fig. 5. Because τ_E is much higher for the multi-step chemistry,
- h_c^{\star} values obtained with 9S18R are higher than those obtained with 3S1R, independently of the temperature. For relatively low pressure (p = 5atm), the critical hot spot size is higher than half an obstacle height (h) and tends to h as the temperature becomes close to the temperature threshold. Increasing the pressure leads to a radical decrease of h_c^{\star} to meet values close to the one
- ²⁷⁰ obtained with the single-step mechanism. This implies that, for this canonic scenario and when using detailed hydrogen/air schemes, conditions where energy can be fed to pressure waves can only be obtained when strong shocks

are involved, resulting in drastic pressure increase. This conclusion was also reached by Ivanov *et al.* [2] and Liberman *et al.* [40] who computed the hot

- ²⁷⁵ spot problem with a more detailed hydrogen/air reaction scheme. Note that, for the single-step mechanism, conditions where HS can lead to an increase in pressure waves amplitude can be met even for temperatures lower than the ignition threshold ($T^* < 0$). Fig. 5(Right) shows that for the 3S1R case, there exists a temperature gradient for which DDT can occur for a large temperature
- ²⁸⁰ range, and for conditions below the ignition threshold. Increasing the hot spot peak temperature results in an increase in the minimum temperature gradient. On the other hand, the multi-step scheme allows DDT for high temperatures only ($\geq 1100 \ K$). As the pressure increases, the DDT region tends towards higher temperatures also pointing out the need for strong shocks. Note that the ²⁸⁵ single-step mechanism shows almost no pressure dependance.
- The hot spot problem may represent a simplified scenario far from the realistic conditions that lead to DDT in gas explosions. In practice, the hot kernel is certainly non-spherical and the distribution of the temperature gradient can be heterogeneous. In addition, reactants mixing can occur leading to additional
- reactivity gradients. However, Fig. 5 suggests that the single-step approach allows DDT to occur for conditions far less restrictive than those required by the detailed hydrogen/air chemistry. The latter, which is validated against experimental data, may not even allow DDT via gradient of reactivity in configurations such as the BR30hS300 channel, unless strong shocks are formed.
- Further investigation is needed to assess the extent of such conclusions to other mixtures and fuels.

4. Numerical simulation of the BR30hS300 configuration

4.1. Flame speed and DDT

The conclusions of the previous section are assessed on a more complex sce-³⁰⁰ nario of DDT in a confined and obstructed channel. The explosion scenario of the BR30hS300 [26] is computed using the single-step and multi-step mechanisms. The evolution of the non-dimensional flame tip velocity s_{tip}/D_{CJ} versus the flame tip position is displayed in Fig. 6(left). The flame tip position is defined by $c = Y_{H_2}/Y_{H_2,u} = 0.95$ for the detailed chemistry and $c = Y_{S_1}/Y_{S_1,u} = 0.95$

- for the single-step mechanism, where u denotes unburned gases. The numerical simulations correctly capture the main features of the experiment. Upon ignition, the flame undergoes multiple accelerations, which are well reproduced by both chemical mechanisms. This can be explained by the fact that 3S1R and 9S18R share the same laminar flame speed s_L and burnt gas temperature
- T_b , which allows a good prediction of reactants consumption and gas expansion during flame propagation. In addition, according to Joulin and Mitani [41] and Sun *et al.* [42], the effective Lewis number Le_{eff} of two reactants flames depends on the reaction order, the equivalence ratio and the Lewis number of the reactants. Le_{eff} reduces to the lewis number of the fuel (oxydizer) in the case
- of very lean (rich) flames. In between, a gradual evolution of Le_{eff} is observed. At stoichiometry, Le_{eff} is close to unity for hydrogen/air flames, which: 1) depletes thermo-diffusive instabilities (confirmed by the experiment); 2) makes the flame speed response to stretch identical for both chemistries.
- Before the first obstacle, FA is laminar in nature and is induced by the strong channel confinement (high S/H ratio). The latter quickly forces the flame to transition from the spherical to the finger shape with a rapid increase in flame surface as seen in Fig. 6(right). The obstacles ahead of the flame enhance FA with successive and stronger flame surface increases, until a sharp increase in flame speed is observed, indicative of a transition from deflagration to detonation.

While both mechanisms predict DDT, its location is strongly impacted by kinetics. Nevertheless, DDT was observed in both simulations just one section either before (9S18R) or after (3S1R) the one predicted by the experiments. Prior to DDT, both flames have reached the fast propagation regime: s_{tip} is

equal to $0.4D_{CJ}(0.5D_{CJ})$ for the 9S18R (3S1R respectively) case. The value $D_{CJ}/2$ is equal to the CJ deflagration speed and corresponds to the maximum deflagration speed [15]. Figure 6 shows that, in the 3S1R case, the flame accel-



Figure 6: Evolution of the non-dimensional flame tip speed (left) and nondimensional flame surface (right) with the flame tip position. The gray area represents the experimental shots. The vertical bars represent the position of the seven obstacles.

eration process slows down when this limit value is reached $(X \in [1.45, 1.75]m)$. The flame surface also reaches a plateau at this regime. After the abrupt DDT

process, and in accordance with experimental data, the flame propagation speed reaches a quasi-steady value close to the predicted autonomous detonation speed D_{CJ} . The non-dimensional flame surface A_f/H then quickly relaxes to unity showing that a quasi-planar detonation propagates in the channel.

The flame acceleration phase, as a necessary step for creating the appropriate conditions for DDT, is briefly analyzed in Section 4.2. Then, the flow conditions prior to DDT are investigated in details in Section 4.3 to determine whether the ingredients that lead to detonation are identical for both chemistries.

4.2. FA phase

Few microseconds after ignition, the flame undergoes the first acceleration episodes due essentially to confinement (finger flame [6]). Starting from the first obstacle, additional FA mechanisms are triggered. The first one is related to the sudden flow contraction. The fresh gases, pushed by the reactive piston, are accelerated when they penetrate the congestion. When the flame starts interacting with the obstacle, the jump velocity condition induces a sudden increase of the flame speed. This mechanism is however limited to the vicinity of the ob-



Figure 7: Numerical Schlieren $(\nabla \rho / \rho)$ and isocontour of progress variable c = 0.5 showing the flame interaction with the first obstacle. Left: single-step mechanism. Right: Multi-step mechanism.

stacle. The FA process continues downstream of the first congestion because of the flame interaction with the flow structures. When the tip of the combustion wave exits the congestion, the remaining part of the reactive surface journeys in a turbulent flow (where vortex shedding has been triggered earlier). Conse-

quently, the flame front is wrinkled as illustrated in Fig. 7 for both chemistries. Flame vortex interactions account for the strong increase of the flame surface just downstream of the congestion observed in Fig. 6(right). Because of the large obstacle spacing to channel height ratio (S/H = 5), the contribution of the congestion to the flame acceleration stops a certain distance away from the obstacle and the flame surface drops significantly. This explains the repeated flame acceleration-deceleration phases observed for both chemical mechanisms in Fig. 6.

Contrary to the deflagration-precursor shock structure observed in smooth channels [8, 20, 23], repeated shock reflections at the obstacles result in an ³⁶⁵ unstructured shock network ahead of the flame (Figs. 8 and 9). When leaving the second obstacle, the flame is highly turbulent as seen at t = 8.10 ms(t = 6.72 ms) for the single-step (multi-step) mechanism respectively.



Figure 8: Numerical Schlieren $(\nabla \rho / \rho)$ and isocontour of progress variable c = 0.5 showing the shocks network formed ahead of the flame. RM denotes the Richtmyer-Meshkov (RM) instability. Left: single-step mechanism. Right: Multi-step mechanism.

Shock reflections at the obstacles allow flame-shock interactions. This is highlighted in Fig. 9 in the 3S1R case where a Richtmyer-Meshkov (RM) instability is triggered with a large funnel of unburned material penetrating the region of burnt gases. This instability further contributes to the increase of the flame surface. Until the fourth obstacle, FA in both simulations matches reasonably well. The reason why DDT is observed first with the detailed chemistry and delayed with the reduced mechanism is discussed in the next section.

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4.3. Detailed analysis of DDT onset

The detonation onset location is strongly impacted by the chemistry modeling as shown in Fig. 6. There may be two possible explanations for this: 1) the DDT mechanism is the same but the necessary conditions to trigger it are met earlier in the 9S18R case than the 3S1R case; 2) the mechanism is different and

involves different ingredients to initiate detonation. A first step towards identifying the key elements responsible for DDT in both simulations is to look at the



Figure 9: Numerical Schlieren $(\nabla \rho / \rho)$ and isocontour of progress variable c = 0.5 showing the highly turbulent flame crossing the third obstacle. RM denotes the Richtmyer-Meshkov (RM) instability. Left: single-step case. Right: multi-step case.



Figure 10: Zoom on the onset of detonation. Pressure field and isocontour of progress variable c = 0.5 are displayed. The dashed lines correspond to a cut through which DDT is analyzed in Sections 4.3.1 and 4.3.2. Left: single-step case. Right: multi-step case.

DDT location. Figure 10 displays the pressure field when the detonation wave starts to form. In both cases, DDT occurs close to the obstacles, in regions of

highly compressed gas. In the 3S1R case, DDT starts in the unreacted mixture ahead of the deflagration front. On the other hand, DDT seems to take place either in the flame brush or between the flame and the wall for the 9S18R case. Figure 10 suggests that the deflagration may play a role in the DDT scenario

with 9S18R.

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- Figures 11 and 12 focus on the moments surrounding DDT for the single-step and the multi-step mechanisms respectively. For the 3S1R case, an Incident Shock (IS) gets reflected at the sixth obstacle (X = 1.744 m) and a Compression Wave (CW) is seen coming in the opposite direction to the reflected shock (RS). The collision between CW and RS leads to the formation of a highly com-
- ³⁹⁵ pressed and heated region (HCG) from which a detonation wave emerges a few microseconds later. A detailed analysis of the detonation onset in this case is performed in Section 4.3.1. Shock reflection is also involved in the DDT process in the detailed chemistry case but the scenario is completely different. Figure 12 shows that, as a consequence of the reflected shock crossing the flame path, a



Figure 11: Key events surrounding the detonation onset for the 3S1R simulation. Pressure gradient ∇P is displayed. x_w represents the distance from the obstacle 6 left wall (grey dashed area). IS is the incident shock. RS and CW indicate the reflected shock and a compression wave respectively. HCG stands for Highly Compressed Gas. A detonation wave is triggered at t = 10.561ms and propagates along the obstacle.

⁴⁰⁰ Pressure Pulse (PP) forms at the flame front and leads to detonation initiation inside the flame brush. This confirms that the deflagration is indeed an active participant in the DDT mechanism observed for the 9S18R case. The formation of the pressure pulse and the exact mechanism behind the sudden transition in propagation regime inside the flame front are analyzed in Section 4.3.2.

405 4.3.1. Single-step chemistry and the Zeldovich mechanism

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DDT in the 3S1R case takes place in the unreacted mixture as already observed in other simulations [1]. The detonation wave is initiated behind the reflected shock observed in Fig. 11. To understand the effect of this shock on the gas and its role in the DDT process, the simulation is inspected through the cut line presented in Fig. 10(Right), positioned $500\mu m$ from the bottom wall. The result is displayed in Fig. 13.

The propagation of IS, RS and CW, shown in Fig. 11, can be observed in Fig. 13(a,b). As a result of the incident shock reflection on the obstacle, a pressure and temperature increase can be observed behind RS. The latter can interact back with the boundary layer and a further increase of temperature is



Figure 12: Key events surrounding the detonation onset for the 9S18R simulation. Pressure gradient ∇P is displayed. x_w represents the distance from the obstacle 4 right wall (gray dashed area). The deflagration front is represented by the isocontrour of progress variable c = 0.5. IS and RS denote the incident shock and the reflected shock respectively. PP is the pressure pulse observed inside the flame brush prior the detonation onset. D represents the detonation wave.

observed due to viscous heating. A region of hot unburned gas $(T \ge 750K)$ can then be observed at the RS tail. The collision of the reflected shock with a compression wave propagating in the opposite direction leads to the formation of a region of hot $(T \ge 800K)$ and highly compressed $(P \ge 35bar)$ unburned

- reactants called Hot Spot (HS). A detonation wave clearly emerges from HS as a strong shock (see Fig. 13(a)) propagates away from HS, coupled to a reaction wave suggested by the formation of combustion products behind it in Fig. 13(c). A retonation wave can be observed in Fig. 13(a) as a result of the reflection of the detonation front on the obstacle. The conditions in pressure and tempera-
- ⁴²⁵ ture observed inside HS are caused by the strong leading shock (IS) reflecting off the wall. This explains why DDT was delayed until the sixth obstacle in this case, allowing the leading shock to gain in strength while the flame propagates in the channel.

To understand why a detonation wave emerges from the hot spot, the evolution of the variables ε and ξ is displayed in Fig. 14. The formation of the hot unburned region HS coincides with values of ε higher that unity. This means that in HS, a compression wave can form and gain in amplitude as its residence time in HS is higher than the time needed locally for the mixture to deliver chemical energy. On the other hand, in the hot spot, for $T \leq 800K$, high values

- of ξ are met, since the gradient $\partial \tau_I / \partial T$ is high for low values of temperature. 435 The increase in temperature inside HS leads to higher values of energy input to pressure waves $\varepsilon \geq 5$ just prior to DDT and lower values of ξ . This means that the evolution of HS was towards high energy transmission to pressure waves $(\varepsilon \nearrow)$ and resonance between the latter and the reaction fronts that emerge
- from HS $(\xi \to 1^+)$. The initiation of the detonation wave in the case 3S1R is 440 therefore the result of the classical Zeldovich mechanism. Note that the temperature conditions met inside the hot spot would have never led to detonation initiation via the Zeldovich mechanism if the multi-step mechanism is used. Indeed, the flame residence time inside an obstructed section can
- be defined as $\tau_f = S/(D_{CJ}/2) \approx 300 \mu s$. Therefore, to allow ignition at these 445 conditions in pressure (i.e. $\tau_I \leq \tau_f$), the minimum temperature inside the hot spot should be higher that 1100K, which would have required a much stronger shock IS than the one observed in the 3S1R simulation.

4.3.2. Detailed chemistry and pressure pulse amplification

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- The procedure proposed in Section 4.3.1 is used to study DDT in the 9S18R case. The computation is investigated through the cut line shown in Fig. 10(Left). Because the detonation does not start from a hot spot, the variables ε and ξ are not suited for the 9S18R simulation. However one can propose a generalized form for $\varepsilon - \xi$ that takes into account the distinct scenario observed in the 455 multi-step simulation.
 - A pressure wave that crosses a flame front gains in amplitude due to the presence of heat release [43]. To measure the energy input by the flame to the pressure wave, the variable $\varepsilon_f = (\delta_L/a)/\tau_{H_{RR}}$ is introduced, where $\tau_{H_{RR}} = \rho Q/H_{RR}$, with H_{RR} the heat release rate and δ_L the flame thermal thickness. ε_f compares
- the residence time of a pressure wave inside the flame front to the time neces-460 sary for the chemical energy to be released inside the combustion wave. Like $\xi, \xi_f = s_{tip}/(u+a)$ is a velocity ratio. ξ_f compares the flame tip propagation



Figure 13: Time evolution of pressure (a), temperature (b) and $\lambda = 1 - c$ (c) in the single-step case through the cut line presented in Fig. 10(Right), positioned 500 μm from the bottom wall. x_w is the distance from the obstacle 6 left wall. IS, RS, CS, R and D denote an incident shock, a reflected shock, a compression wave, the retonation wave and the detonation wave respectively. HS is a hot spot.



Figure 14: Time evolution of temperature (a), ε (b) and ξ (c) in the single-step case through the cut line presented in Fig. 10(Right), positioned 500 μ m from the bottom wall. x_w is the distance from the obstacle 6 left wall. IS, RS, CS, R and D denote an incident shock, a reflected shock, a compression wave, the retonation wave and the detonation wave respectively. HS is a hot spot.



Figure 15: Time evolution of the pressure gradient (a), ε_f (b) and ξ_f (c) for the multi-step case through the cut line presented in Fig. 10(Left). y_w is the distance to the bottom wall. RS and D denote the reflected shock and the detonation wave respectively. The black contours indicate the flame tip and tail. The gray contours in (b) are isolevels of temperature in the unburned region in front of the flame front.

velocity to the pressure wave propagation velocity.

- ε_f and ξ_f are displayed in Fig. 15 along with the pressure gradient. The reflected shock RS shown in Fig. 12 appears crossing the flame front. The unburned material, in front of the flame front, is compressed and heated as a result of consecutive shock reflections between the wall and the flame front. In Fig. 15(b), the grey contours indicate that the temperature of the reactants increases to reach temperatures slightly higher than 750K. This temperature,
- ⁴⁷⁰ however, is not sufficient to trigger DDT in the unreacted material. Instead another mechanism takes place. The interaction of RS with the flame front has two consequences on the flame front: 1) the formation of a pressure pulse PP inside the reaction zone; 2) the slowdown of the latter due to a backflow of fresh gases (see Fig. 15(c) and [43]). From $t \approx 8.007 \text{ ms}$ and the formation of
- ⁴⁷⁵ PP, an $\varepsilon_f \xi_f$ analysis allows to explain the detonation initiation. At first PP propagates in a region where ε_f is lower than unity. PP cannot gain enough



Figure 16: 1D stoichiometric hydrogen/air premixed flame structure at 5 bar and 300 K. Y_k denotes the mass fraction of species k.

energy and its amplitude decreases as PP propagates towards the flame tail (black arrow in Fig. 15(a)). However because of the flame speed stagnation, the PP propagation velocity is just slightly higher than the flame front propagation

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- speed which prevents PP from exiting the reaction zone and forces it to gain in amplitude. As seen in Fig. 15(b), a process of pressure pulse amplification is therefore triggered since ε_f progressively increases to reach values close to unity. At this stage ($t \approx 8.008 \text{ ms}$), a sudden flame acceleration is observed which prevents PP from exiting the reaction zone. This situation forces PP to further gain in amplitude. DDT inside the flame brush is then triggered via
- ⁴⁸⁵ further gain in amplitude. DDT inside the flame brush is then triggered via shock wave amplification by flame-acoustics coherence (a form of the SWACER mechanism [15]).

This transition to detonation by pressure pulse amplification was already observed by Goodwin et. al. [22, 44] who used a single-step chemical scheme. While this mechanism is not specific to multi-step mechanisms, it is often observed to be the trigger of detonation initiation in the case of detailed chemistry

modeling [2, 43, 45]. The reason behind it is first related to the difficulty to

meet the necessary conditions for the Zeldovich mechanism to trigger DDT as long as strong shocks are not formed, as explained in Section 3.4.2. The second

- reason can be attributed to the presence of highly reactive radical species in the multi-step mechanisms, such as HO_2 . Figure 16 displays the 1D mass fraction profile of radicals as well as the heat release rate profile in the progress variable space for P=5bar, which is close to the pressure condition prior to DDT in the 9S18R case. HO_2 initiates the first exothermic reactions responsible for the heat
- release inside the flame, before H_2O_2 and later H take over. In addition, HO_2 is able to deliver chemical energy at small length scales and can explain the formation of the pressure pulse PP. Indeed, Fig. 17b displays the non-dimensionnal HO_2 mass fraction and shows an overshoot in these radical species as soon as RS starts interacting with the flame front suggesting a strong shock-radical
- species interaction to form PP. The chemical energy delivered by HO_2 can be measured by introducing $\varepsilon_{HO_2} = (\delta_{th}/a)(\dot{\omega}_{HO_2}/\rho)$ where $\delta_{th} = D_{th}/s_L$ is the flame preheat region thickness, D_{th} being the thermal diffusivity, and $\dot{\omega}_{HO_2}$ the radical species source term. ε_{HO_2} measures the chemical energy input by the radical species HO_2 into a pressure wave during its residence inside the preheat
- region of the flame. ε_{HO_2} is displayed in Fig. 17(c) and shows that the small time-scales associated with $\dot{\omega}_{HO_2}$ result in a rapid energy input in the small preheat region. The subsequent positive feedback between HO_2 and the reflected shock wave leads to the formation of PP.

5. Conclusion

- The influence of chemistry modeling on the intricate Deflagration to Detonation Transition (DDT) problem is investigated through two chemical mechanisms: 1) a single-step mechanism (3S1R) designed to reproduce the key features of flame and detonation propagation; 2) a multi-step mechanism (9S18R) validated against experimental data in terms of flame speed for a large range of
- ⁵²⁰ equivalence ratios and in terms of ignition delays for a large range of temperature.



Figure 17: Time evolution of the pressure gradient (a), HO_2 mass fraction (b) and $\varepsilon_{HO_2} = (\delta_{th}/a)(\dot{\omega}_{HO_2}/\rho)$ (c) for the multi-step case through the cut line presented in Fig. 10(Left). $[Y_k]_{max}$ is the maximum species mass fraction inside the post-processing window. y_w is the distance to the bottom wall. RS and D denote the reflected shock and the detonation wave respectively. The black contours indicate the flame tip and tail.
First the idealized hot spot problem with constant temperature gradient is considered. The requirements in terms of temperature and radius for a hot spot to trigger DDT are assessed based on the Bradley et al. [39, 46, 47] analysis.

- Results show that the single-step approach can lead to detonation onset from a 525 hot spot for conditions of temperature much lower that the ignition threshold obtained from the experimental data. On the other hand, with 9S18R, the hot spot sizes required for pressure wave amplification by chemical energy input are much higher than those obtained with 3S1R for relatively low pressure con-
- ditions, and become comparable at high pressure. This finding suggests that 530 when using a detailed mechanism, the necessary conditions to allow detonation initiation via gradient of reactivity can hardly be met as long as strong shocks are not formed. This is true for hydrogen/air combustion, further studies however are needed to investigate the extent of such a conclusion to other fuels and mixtures.

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The DDT problem was also studied through a more realistic configuration: the DDT experiment of the Gravent database [26] where a gas explosion propagates in a confined and obstructed channel filled with a stoichiometric hydrogen/air mixture. Shock reflections were found to be the common ingredient for DDT

- for all simulations. However, a great influence of the chemistry modeling on the DDT process was observed. In the 3S1R case, the detonation wave emerges from a region of unreacted materiel characterized by a gradient of reactivity, consistently with the Bradley et al. [39, 46, 47] $\varepsilon - \xi$ analysis. High energy input and resonance between combustion wave and compression wave are required to
- trigger a detonation. On the other hand, the transition to detonation is found 545 to be triggered inside the flame brush in the 9S18R case. A generalized form of the $\varepsilon - \xi$ variables of Bradley is proposed to understand DDT in this case. A pressure pulse (PP) amplification process is initiated inside the flame front. DDT is then attributed to the resonance between PP and the flame tip and to
- the high chemical energy delivered to PP by the exothermic region inside the 550 flame front. There are reasons to believe that this mechanism might be more frequent when hydrogen/air complex chemistry is considered: 1) the necessary

conditions for a hot spot to lead to detonation wave formation may be difficult to meet when using detailed chemistry; 2) highly reactive species, like HO_2 , are found to be the trigger for the pressure pulse formation, which is the first step

of the pressure pulse amplification process.

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Figure 9.1: Pressure field history during the detonation front propagation in the unobstructed region of the channel, i.e. just after the last obstacle. (Left) single-step chemistry. (Right) multi-step chemistry. Multiple fields are superimposed to show detonation front dynamics. The time separating each frame in the multi-step case is two times higher than the single step case.

9.2 The detonation front

After the abrupt DDT process, a detonation wave forms. In this section, the detonation front structure is investigated. In Fig. 9.1, snapshots of the detonation front during its propagation in the unobstructed region of the channel are superimposed. Even though the detonation speed is reproduced with reasonable agreement, a strong influence of the combustion mechanism on the detonation structure is observed.

In the single-step case, the detonation front propagation is characterized by a transverse instability, leading to the formation of a cellular pattern following the leading shock path. The detonation cell average width 10.2 mm is close to the experimental value $\approx 8.7mm$, which is expected since the 3S1R mechanism is designed to reproduce the correct detonation thickness, hence the dimensions of the diamond pattern. Surprisingly, the detonation front is stable in the 9S18R case. An explanation for the stable nature of the detonation front in the 9S18R case is proposed in the following.

In the 3S1R case, triple points are visible at the point of intersection between the transverse and leading shock waves (Fig. 9.2(left)). A further focus on the detonation front in (Fig. 9.3) shows that its structure, in this case, is as expected: A shock initiates the Von-Neumann state (VN) followed by an exothermal reaction zone. When this structure is taken into account, multidimensional instability analysis confirms the existence of unstable modes explaining the diamond pattern. The key element is the sensitivity of the induction time to fluctuations in the VN state temperature, hence the length of the induction zone downstream of VN. A coupling between the heat release rate and the entropy-vorticity wave controls the instability in the case of a strongly overdriven detonation Clavin et al. (1997), whereas near the self-sustained detonation regime the role of the entropy-vorticity wave vanishes and the instability results from the coupling between the acoustic wave and the heat release rate Clavin and Williams (2009). The diamond pattern dimensions



Figure 9.2: Non-dimensional density ρ/ρ_u field across the detonation front. (Left) single-step chemistry. (Right) multi-step chemistry. The arrows highlight the transverse shock trajectories. TP denotes a triple point.

can be estimated using a pulsation time of the order of the induction time at the VN state, τ_{VN} Clavin and Searby (2016). The transverse and longitudinal disturbances travel at the sound speed at the VN state a_{VN} and the detonation speed D respectively. The detonation cell dimensions would be $a_{VN}\tau_{VN} = 11.9mm$ and $D\tau_{VN} = 20.4mm$, which are close to the lengths found in the simulation, i.e. 10.2mm and 26mm respectively.

The detonation structure in the 9S18R case however is different from the classical ZND structure:

- the shock present in the 9S18R case is weak $(\rho_1/\rho_u \approx 2.5)$ compared to the one shown in the 3S1R case $(\rho_1/\rho_u \approx 10 \text{ very close to the VN state})$, the index u and 1 denoting the pre-shock and post-shock conditions respectively. The value ρ_1/ρ_u given by the 9S18R mechanism is closer to the CJ condition $(\rho_{CJ}/\rho_u = 1.8)$. Figure 9.4(left) shows profiles of temperature and pressure for the 9S18R case through the line Y = H/2 = 30 mm for five different instants. A steady jump condition across the detonation is observed, in fair agreement with the CJ to fresh pressure and temperature ratios.
- a zoom on the detonation structure in the 9S18R case, displayed in Fig. 9.4(right), shows that the fresh mixture is gradually brought to the CJ state without going through the VN state. Fuel consumption is initiated at the detonation tip and is terminated at the locus of the sonic plane (M = 1), which coincides with the CJ plane.

The same detonation profile shown in Fig. 9.4(right) is analyzed in the progress variable space and in the $\left(\frac{P}{P_u} - \frac{\rho_u}{\rho}\right)$ plane in Fig. 9.5. Figure 9.5(left) shows that the pressure increase across the detonation front proceeds with chemical heat release and fuel consumption. The position of the maximum pressure value coincides with the position of



Figure 9.3: Non-dimensional density ρ/ρ_u field across the detonation front with contours of progress variable c indicating the region of fuel consumption. (Left) single-step chemistry. (Right) multi-step chemistry.

the peak heat release rate H_{RR}^{max} . The unconventional nature of the detonation structure in the 9S18R case is also confirmed in Fig. 9.5(right), where it is compared to the ZND structure. A clear bypass of the VN state is observed and the burnt gas state is directly reached from the initial state. However, in this case, one could expect to observe a solution following a straight line from the fresh to the burnt gases. Surprisingly this is not the case and a curved line is observed. This slight deviation from the straight line is attributed to the varying γ through the solution. The unstable nature of detonations, nears the CJ limit, results from the perturbation of the heat release rate profile behind the leading shock, when the velocity of the latter is perturbed. This mechanism does not hold when the ZND structure is not present, as it is the case for the 9S18R detonation. This unconventional solution was already mentioned by Yu et al. Yu and Chen (2015) in one-dimensional DDT computations using detailed chemistry for the same mixture considered in this paper (i.e. stoichiometric hydrogen/air). They showed that the detonation do not correspond to the ZND structure. However, in their case, the fresh mixture ahead of the detonation front is in an ignitable state $(T_u \geq 1000K \text{ and } P_u \geq 20 \text{ bar})$, which could explain the initiation of the chemical reactions before the shock. In the present case, the fresh mixture is in a frozen state $(T_u \approx 355K \text{ and } P_u \approx 2 \text{ bar})$ and the activation of chemical reactions can not occur before the leading shock since the time scale of elastic collisions controlling the inner structure of inert shocks (small number of elastic collisions) is smaller that the time scale of the heat release rate, as argued by Clavin and Searby (2016).

The only explanation left for the unusual detonation structure observed in the 9S18R case is related to numerical artifacts and the method used to handle shocks in AVBP. Indeed, the Cook and Cabot (2004) artificial viscosity is used in this thesis. The activation of the associated sensor and its consequences on the detonation structure are highlighted in Fig. 9.6, where the artificial bulk viscosity k' (Eq. (2.41)), used to smooth shocks, is



Figure 9.4: Left: Non-dimensional pressure P/P_u (top) and non-dimensional temperature T/T_u (bottom) profiles across the line $Y = H/2 = 30 \ mm$ for the 9S18R case. The time separating two instants is constant and equal to $10 \ \mu s$. Right: zoom on the detonation profile showing P/P_u , T/T_u and the flow Mach number M through the detonation front. M is computed in the reference frame of the shock.



Figure 9.5: Left: Non-dimensional pressure P/P_u , temperature T/T_u , heat release rate H_{RR}/H_{RR}^{max} together with the Mach number M profile as a function of the progress variable 1-c. This corresponds to the instant shown in Fig. 9.4(right). Right: $\left(\frac{P}{P_u} - \frac{\rho_u}{\rho}\right)$ plane for the 9S18R detonation confronted to the one given by the ZND structure.

displayed. The consequent artificial thickening of the leading shock may be sufficient to observe the activation of chemical reactions, associated with highly reactive species like HO_2 before the VN state is reached. Chemical reactions then bring the gas directly to the burnt gas state (bypassing the VN state). While the Cook and Cabot (2004) artificial viscosity is also used in the 3S1R mechanism, the thickening of the leading shock does not interfere with the ZND structure in the 3S1R case. Indeed, the high activation energies used in one-step mechanisms introduce a cut-off temperature, below which the chemical source term is negligible. Therefore, in the 3S1R case, chemical reactions can not proceed in the artificial shock thickness and the heat release is initiated behind the leading shock.

To conclude, the methodology to handle shocks used in AVBP seems to interfere with the detonation structure in the multi-step mechanism, leading to an unusual detonation structure. Further investigations are needed to understand the emergence of this structure. A first step towards the understanding of this peculiar behavior is to perform numerical simulations of detonation propagation (i.e. via direct detonation initiation). This simpler



Figure 9.6: Profiles of non-dimensional pressure P/P_u , temperature T/T_u and artificial bulk viscosity k' (Eq. (2.41)) normalized by the shear viscosity μ . This corresponds to the instant shown in Fig. 9.4(right).

problem can highlight the different numerical issues in handling detonation fronts. It should also highlight the requirements in terms of grid resolution to adequately reproduce the key features of the detonation.

Part III

What is the underlying mechanism behind some solid flame inhibitors?

Mitigation of vapour cloud explosions

Vapour cloud explosions may occur in all types of installations and environments. The damages inflicted to buildings and civilians vary from moderate to severe depending on multiple factors: ignition source, type of flammable material, levels of confinement and congestion on site, regime of combustion wave propagation, etc... The implementation of preventive measures has led to a significant decrease of the occurrence of such disasters. However, when they do happen, VCE's are tragic events that result in a high number of injuries and fatalities in addition to billions of dollars of world-wide losses to mining, process and energy industries. As often, they are triggered by a cascade of events that can hardly be predicted. Take the Buncefield accident for example. The official report, delivered by the british Government, revealed that a switch failure caused an overflow of unleaded petrol into a tank. An alarm was designed to activate in case of switch failure but it failed too. The combination of these two malfunctions was so unlikely that no procedure was implemented to deal with this scenario, which led to the formation of a large cloud of flammable mixture. At that point, any energy source sufficiently high to trigger chemical reactions can give rise to a large scale explosion. Chapters 7-9 showed that a combination of confinement and congestion, two ingredients commonly found on industrial sites, can result in levels of overpressure high enough ($\geq 4 \ bar$) to heavily damage most of the process installations, even when no detonation is initiated. The study of past accidents highlights the need for mitigative procedures that can reduce the effects of explosions in case of failure of the preventive measures.

As a prelude to discussing the different techniques that can be used to mitigate VCEs, it is useful to provide a definition of the different mitigation strategies. A distinction is made (Linteris (2004)) between two effects on the flame:

- The flame suppression (extinguishement, extinction) refers to the case where the flame is weakened to the point where it can no longer stabilize in the flow field.
- The flame inhibition describes the reduction of the overall exothermic reaction rate in the flame, without the intention to suppress the flame.

These two effects can be achieved via thermal (cooling) or chemical mechanisms, or a combination of both. A dominantly physical effect can be achieved by water injection, for example, as discussed in Section 10.1. A more effective way to deal with flames, involving a profound effect on flame chemistry, is described in Sections 10.2 and 10.3. Section 10.4 identifies the conditions required to mitigate VCEs.

10.1 Physical mitigation using water sprays

One of the first techniques to be considered was inspired from fire safety and involves water sprays. This method was tested on deflagrations by a number of investigators (Carlson et al. (1973); M. et al. (1977); Zalosh and Bajpai (1982); Acton et al. (1991); Thomas et al. (1991); van Wingerden and Wilkins (1995); Bjerketvedt et al. (1997); Van Wingerden (2000); Thomas (2000)) with satisfactory results. During these experiments, a significant pressure peak reduction was observed depending on the droplet size distribution (up to 75% in Acton et al. (1991), and $\approx 70\%$ in Van Wingerden (2000)). Thomas (2000) emphasized the role of the small droplets (smaller than some critical diameter d_p^c that may depend on the flame and the flow) in a polydispersed phase. These droplets act as a heat sink either during the evaporation of the particle inside the flame or via the dilution of the mixture when the particle is preheated and evaporates before reaching the flame front as indicated by M. et al. (1977); Zalosh and Bajpai (1982).

Droplets larger than d_p^c require an evaporation time larger than the time scale of flame propagation. As a result they will not have an impact on the explosion. Simple analytical calculations (M. et al. (1977)) showed that individual droplets must be of the order of only few tens of microns in diameter. Despite this observation, sprays with large droplet size distributions (of the order of hundreds of microns to millimeters) have been reported to be effective as well (see Thomas et al. (1991); van Wingerden and Wilkins (1995)). The reason why these large droplets are able to reduce the pressure peak is related to the particle secondary break-up mechanism. Indeed, the piston induced flow of fresh gases can break large droplets into smaller ones, typically smaller than d_p^c , if the hydrodynamic forces applied on the particle surface are larger that the force holding the droplet together, namely the surface tension. Because the resulting droplets belong now to the class of particles able to evaporate inside the flame, this results in a successful mitigation. As shown in Chapters 7-9, deflagrations can produce the ingredients able to trigger this secondary break-up mechanism: 1) sudden accelerations of the fresh gases; 2) hight turbulent fluctuations; 3) strong shocks. This mechanism is controlled by the Weber number W_e , defined by the ratio of the inertial force on the droplet surface to its surface tension:

$$W_e = \rho d_p \|\mathbf{u}_p - \mathbf{u}\|^2 / \sigma_{surf} \tag{10.1}$$

where **u** and ρ are the fluid velocity and density respectively. **u**_p and d_p are the particle velocity and diameter respectively. σ_{surf} is the particle surface tension. Generally,

- for very low Weber numbers, namely $W_e < 10$, no break up occurs. This is the case for particles in the range $[10, 200]\mu m$ in the experiments of van Wingerden and Wilkins (1995). The flow is unable to rupture these droplets, which are too large to evaporate inside the flame.
- For larger Weber numbers, rupture of the droplet surface occurs, thereby forming much smaller droplets. Depending on W_e , and other parameters like the Ohnesorge number (see Jain et al. (2015)), this rupture exhibits different modes as illustrated in Fig. 10.1.

The necessity to take into account this secondary break-up phenomenon lies in the fact that these larger droplets generally contain most of the water mass as highlighted by Thomas (2000); Chauvin et al. (2015); Jarsalé et al. (2016). When they rupture, they can



Figure 10.1: Secondary droplet break up for two increasing Weber numbers $W_e = 20, 120$. From Jain et al. (2015)

provide a great amount of fine droplets that will contribute to the heat sink.

Water sprays have also been tested on detonations. The successful quenching of detonations has been reported by Gerstein et al. (1954); Thomas et al. (1990). Water sprays have also been used to either delay Boeck et al. (2015) or suppress Gerstein et al. (1954) DDT. As pointed out by Gerstein et al. (1954) and Boeck et al. (2015) the amount of water needed to either quench a detonation or suppress DDT can be huge. For lower quantities, only a small detonation velocity deficit has been observed (Boeck et al. (2015); Jarsalé et al. (2016)). This represents the main drawback in using water sprays. A more efficient technique, per mass basis, can be obtained by taking advantage of the chemical nature of these explosions. This technique is discussed in Section 10.2.

10.2 Dominantly chemical mitigation using metal containing compounds

Identifying more and more candidates as mitigation compounds is motivated by the need to find substances that can induce the desired effect on the flame (suppression or inhibition) with the least amount of injected material possible. This criterion is used to rank inhibitors (Rosser et al. (1963); Hastie (1973); Linteris (2004). Their efficiency is then defined by the quantity needed to either extinguish or to obtain a certain reduction of the consumption speed of a given flame. Halogen (F, Cl, Br ...) containing compounds were among the most widely used inhibitors. Their higher efficiency, per mass basis, than chemically inert particles led to the conclusion that their effect on the flame was not only thermal (i.e. cooling) but also chemical (Belles. and O'Neal (1957)). Many complex chemical mechanisms have been proposed to describe the halogenated flame inhibition

(Dixon-Lewis and Simpson (1977); Safieh et al. (1982); Safieh, H. Y. and Van Tiggelen, P. J. (1984); Vandooren et al. (1989); Hamins et al. (1994b); Yang et al. (1994); Lee et al. (1996); Noto et al. (1998b)). The main drawback of these compounds is their threat to the environment. The Kyoto protocol identified the use of ChloroFluoroCarbon (CFC) and BromoFluoroCarbon (BFC) compounds as potential contributors to the greenhouse effect. For this reason, the research is still active to find replacements for these agents Gann (2004); Linteris (2004). Metal (Fe, Mg, Ca...) containing substances have attracted attention because of their high efficiency: some metals being up to eight times more efficient than the most widely used halogen containing inhibitors. In particular, metal salts (like potassium bicarbonate KHCO₃ and sodium bicarbonate NaHCO₃) have received considerable attention recently. Indeed, well controlled laboratory flame studies (Hamins et al. (1994a); Hamins (1998)) showed that, when injected as very fine powders (namely of the order of the micron), they were more effective than other agents considered to replace BFCs, the effectiveness of $KHCO_3$ being about 2.5 times that of NaHCO₃. However, hydrocarbon flames were reported (Chelliah et al. (2003)) to be insensitive to large metal salts particles (namely larger than 30 μm). These last observations lead to the conclusion that the flame inhibition by metal salts may be governed by two ingredients: 1) the particle size distribution in the powder; 2) the alkali metal, i.e. Na or K for example, composing the salt. A description of the inhibition mechanism by metal salts is needed to understand why these two ingredients are of crucial importance.

10.3 The mechanism of flame inhibition by metal salts

For a long time, investigators like Rosser et al. (1963); Birchall (1970); Iya et al. (1975); Jensen et al. (1979); Mitani (1983); Mitani and Niioka (1984); Hynes et al. (1984); Babushok (1998); Williams and Fleming (1999, 2002); Chelliah et al. (2003); Babushok et al. (2003) favored a gas phase interaction between the flame and the solid inhibitors. The mechanism can be summarized as follows:

- 1. As soon as the particle \mathcal{X}_{solid} enters the preheat zone of the flame, its temperature starts increasing. \mathcal{X}_{solid} denotes the sodium bicarbonate particle for example;
- 2. Once the surface temperature of the particle reaches certain temperature T_{decomp} , thermal decomposition occurs. The decomposition reaction can be roughly described by $\mathcal{X}_{\text{solid}} \longrightarrow \mathcal{I}_g + \mathcal{R}_{\text{res}}$, where \mathcal{I}_g is the gaseous agent and \mathcal{R}_{res} is the reaction residual composed of inert species. \mathcal{I}_g is the NaOH species in the case of sodium bicarbonate. It is important to note that this equation is merely an approximate and that the thermal decomposition can be quite complex, involving multiple intermediate steps and phase changes. This is discussed in Chapter 12;
- 3. the gaseous agent \mathcal{I}_g interacts with the flame radicals. \mathcal{I}_g consumes radical species to produce stable species, thereby reducing the heat release rate of the flame. These radical species can be typically H, OH and O.

This mechanism can explain the flame inhibition process only if the thermal decomposition time scale is smaller than the time scale of the flame propagation. This is illustrated



Figure 10.2: Homogeneous flame/solid inhibitor interaction. The outcome of this interaction depends on the time necessary for the particle to reach the decomposition temperature. If it is smaller than the time scale of the flame propagation, a successful inhibition is achieved. Otherwise, the flame advances as if no particle was present.

in Fig. 10.2. As soon as the particle enters the preheat zone of the flame, the whole interaction can be viewed as a race between the flame propagation and the particle decomposition processes. If the time t_{decomp} necessary for the particle to reach the decomposition temperature is larger than the time scale of flame propagation, then the gaseous agent will be liberated in the burnt gases without any impact on the flame course. Otherwise, the particle liberates \mathcal{I}_g inside the flame where it can influence the combustion reactions and reduce the laminar flame speed to the value $s_L^{\mathcal{I}}$. It is worth noting that this comment implies a stepwise behavior of the flame inhibition process, i.e. either the laminar flame speed is reduced to $s_L^{\mathcal{I}}$ or it remains constant and equal to s_L . This is a simplification of the problem and partial reduction of the flame speed is possible. A word on this point is provided in Chapter 12.

Using this mechanism, one can explain the importance of the particle size and the alkali metal in the flame inhibition process:

- t_{decomp} is an increasing function of the initial particle size and reduces to zero for infinitesimally small particles, so that there is always a limit size below which a metal salt is an efficient inhibitor (i.e. t_{decomp} smaller that the time scale of the flame propagation). This limit size depends on both the inhibitor and the flame (Rosser et al. (1963); Dodding et al. (1970)). In practice, such limit sizes can be too low to be realistically achievable for certain flames as pointed out by Hoorelbeke (2011). For particles larger than the limit size, the flame propagation processes are faster than the thermal decomposition and the powder is ineffective;
- For very fine powders, the degree of inhibition can be defined by the ratio $s_L^{\mathcal{I}}/s_L$. The latter depends:
 - for a given inhibitor, on the amount of gaseous agent \mathcal{I}_g liberated inside the flame, which in turn depends on the quantity of fine powder injected in the



Figure 10.3: Photomicrographs taken by scanning electron microscope for the sodium bicarbonate granules. The picture highlights the complex structure of the sodium bicarbonate. From Alanazi (2010).

mixture. The rate of radical species recombination increases with increasing mass fraction of \mathcal{I}_g , thereby decreasing $s_L^{\mathcal{I}}/s_L$. There is however a limit to the effectiveness of the inhibitor as highlighted by Noto et al. (1998a); Dixon-Lewis and Simpson (1977), which is related to the quantity of available radicals to recombine.

- for a given powder quantity, on the gaseous agent \mathcal{I}_g liberated in the flame and its impact on the flame chemistry. The rate of radical species scavenging is controlled by the alkali metal in \mathcal{I}_g . In the case of sodium bicarbonate and potassium bicarbonate, \mathcal{I}_g is respectively KOH and NaOH, with K and Na the alkali metals. Williams and Fleming (1999) performed one-dimensional flames diluted with the same quantity of either KOH or NaOH. A higher flame speed reduction with KOH was observed, which is consistent with the higher efficiency of the potassium bicarbonate compared to the sodium bicarbonate evidenced experimentally. The difference in efficiency was attributed to the activation of different recombination reactions from an alkali to another.

The gas-phase inhibition mechanism is the subject of extensive investigations, it is not however the only possible mechanism. Mainly two reasons can explain why the literature on the subject is focused on the gas phase inhibition mechanism:

- simple decomposition time calculations Rosser et al. (1963) show that typical inhibitors can indeed fully decompose inside the flame for diameters of the order of tens of microns, which implies that the mechanism is possible without excluding other processes;
- it is much simpler than the surface decomposition mechanism, which describes the chemical reactions between the flame radicals and portions of the metal salts present on the surface of the solid particles. Sodium bicarbonate for example exhibits a crystalline-like structure as shown in Fig. 10.3. As discussed by Krasnyansky (2008), flame radical species can attack the particle on multiple sites where catalytic reactions can occur. He concluded that a coordinated international program of studies is needed to assess the relative importance of these different mechanisms in the global inhibition effect.

In this PhD, the sodium bicarbonate is selected to investigate the flame inhibition process. It has been preferred to other substances because: 1) mechanisms modeling the

interaction of the flame with the gaseous agent NaOH are much less complex than with other metals and are well documented; 2) thermochemistry data for sodium containing species can easily be found in the literature. A validation of the mechanism is described in Chapter 12.

10.4 On the conditions required for VCE mitigation by sodium bicarbonate powder

Metal salts have been used with a very high efficiency to prevent, inhibit and extinguish fires. Their applicability to VCEs, however, is not straightforward. Suppose that the combustible cloud, formed prior to VCEs, has been detected and that the industrial site has been evacuated. An ignition of the mixture, somewhere in the buildings, can initiate a rapid escalation of the explosion hazard, resulting in the complete destruction of the installations, hence millions of dollars of financial losses. The role of the mitigative technique is now clear: reduce as much as possible the damage to the different structures of the industrial plant. The conditions required to fulfill this objective are discussed in the following.

The necessity to avoid flame suppression

Suppose that metal salt powder has been injected in very large quantities, so that the flame is weakened to the point where it can no longer stabilize in a given flow. Upon extinction, the remaining part of the reactive cloud, that could not be burnt by the flame, still represents a threat for the integrity of the buildings. Moreover, the activation of the first procedure can exhaust all the available powder. A second ignition can therefore lead to extreme damage. For these reasons, in the context of VCEs, the quantity of powder injected is a delicate compromise: it should lead to the maximum degree of inhibition possible without extinguishing the flame.

The necessity to reduce the flame heat release rate

Now that flame suppression has been ruled out, the remaining option is to let the flame burn the combustible while reducing the damages that it might inflict on buildings. Consider the flame acceleration case discussed in Chapter 7 and let us give some orders of magnitude of the overpressure reduction needed to induce the smallest possible damage. An overpressure peak of about 4 *bar* was recorded. This is sufficient to heavily damage most of the process installations. A reduction of 95% ($\approx 200 \text{ mbar}$) of this peak overpressure could save blast proof buildings but is still enough to destroy the installations that are not blast proof. As discussed in Section 7.3, the pressure build up inside a building is produced by the overall heat release rate in the reaction zone, which can be efficiently reduced using metal salt powders as explained in Section 10.3.

The necessity to control the powder injection procedure

Up until now, the flame inhibition process has been described in its one-dimensional form. The capacity of very fine sodium bicarbonate particles to reduce the laminar flame speed has been established. However, the objective is to use these powders to inhibit VCEs. One has to take into account the multi-dimensionality of the problem before implementing



Figure 10.4: Impact of the inhibitor spatial distribution on the flame surface. The picture presents two limiting cases: an homogeneous distribution (top), which leads to an overall decrease of the flame consumption speed; and a stratified distribution (bottom), which results in the increase of the flame surface.

any mitigative system based on these powders. Perhaps the most important challenge to solve is to deliver the powder as homogeneously as possible to the whole flame front. The reason why the spatial distribution of the particles is a major issue is provided in Fig. 10.4. Suppose that the powders contain very fine particles, in the sense that their decomposition can be complete inside the reaction zone of the flame. If an homogeneous sodium bicarbonate particle distribution (see Fig. 10.4(top)) is delivered to the flame front, a uniform reduction of the flame consumption speed is obtained. This represents the ideal case. The other limiting case corresponds to a strongly nonhomogeneous distribution of the particles ahead of the flame (see Fig. 10.4(bottom)). The part of the flame surface traveling in the region of strong inhibitor concentration will be slowed down compared to the part traveling in the inhibitor free flammable cloud. The global behavior of the flame surface is therefore comparable to the propagation of a flame in a stratified fuel concentration. As highlighted by Boeck et al. (2014), fuel concentration gradients can enhance the flame acceleration process and even facilitate the onset of detonation.

In practice, it is very difficult to ensure an homogeneous distribution of the inhibitor ahead of the flame. Even in well controlled bomb experiments, perturbances on the flame surface because of the solid particle spatial distribution have been reported. For example, Rosser et al. (1963) commented on some spurious results by saying: 'In the absence of powder the flames were approximately hemispherical in shape; ... The presence of powder resulted at times in irregular flame shapes ... In some cases the flames were very long and highly tilted, probably as a result of non-uniform powder distribution'. Multiple scenarios can explain an inhibitor stratification ahead of the self-propagating flame. For example,

- the time scale of the flame propagation can be too small compared to the particle dispersion time scale, so that the deflagration propagates in a cloud where the particle density decreases with increasing distance from the injector (see Fig. 10.5(a));
- the injection system is activated a long time before ignition of the mixture occurs. As a result, when the deflagration emerges, the powder is no longer in suspension in the flammable cloud. As the flame propagates, the particles, that have settled



Figure 10.5: Sketch of two possible scenarios that can lead to a non-homogeneous inhibitor particle distribution ahead of the flame. a) Injection just prior to flame arrival. b) Injection a long time before ignition.

down, are set into motion, so that the flame interacts with the particles only near the ground (see Fig. 10.5(b)).

10.5 Objective of the present study

The impact of solid inhibitors on the flame propagation is investigated in this PhD with regards to VCEs. As explained at the end of Section 10.3, the study focuses on the sodium bicarbonate. Two major problems, relevant to VCEs, are tackled: 1) the effect of the particle size on the flame inhibition process; 2) the effect of nonhomogeneous particle distributions on the flame surface and the flame propagation speed. The different methods employed to perform the analysis are described in Chapter 11. The results are presented in Chapter 12.

Chapter 11

Computational approach to flame inhibition by sodium bicarbonate

Flame inhibition by sodium bicarbonate involves the decomposition of a solid particle as it enters the flame front and the interaction of the liberated gaseous agent with the flame radical species. Only one fuel has been considered for the simulations presented in Chapter 12, namely methane. The numerical investigation of the interaction of a sodium bicarbonate powder with a methane/air flame requires: 1) a formalism to describe the solid particle motion and temperature; 2) a chemical scheme to model the gaseous agent interaction with the flame chemistry. Both aspects are discussed in Sections 11.1 and 11.2 respectively.

11.1 Lagrangian formalism for the solid phase

To describe the evolution of the sodium bicarbonate solid particles, a deterministic Lagrangian approach, called "DPS" for Discrete Particle Simulation, is employed. Each particle in the inhibitor powder is considered as a moving point, exchanging heat, mass and momentum with the gas, assuming that:

- there is no interaction (including collision) between the particles;
- the particles are initially spherical and remain spherical during their thermal decomposition.

The first hypothesis is related to the low inhibitor concentrations employed in all the simulations presented in Chapter 12. The second hypothesis, however, needs to be discussed. Figure 10.3 shows that sodium bicarbonate particles are far from being spherical. Their shape can be complex with varying aspect ratios, which certainly impacts the heat, mass and momentum transfer between the gas and the particle, thereby influencing the flame inhibition process. The present study must be viewed as a first step towards the understanding of the multidimensional inhibitor powder/flame interaction in fast deflagrations. Simplified models, able to reproduce first order effects, are mandatory to keep the computational cost of such complex simulations reasonable.

Under these assumptions, each solid particle is represented by a point in space whose position $(X_{p,i})_{i=1,3}$, mass m_p , velocity $(u_{p,i})_{i=1,3}$ and temperature T_p are given by the following set of equations, based on a deterministic Lagrangian formalism:

$$\frac{DX_{p,i}}{Dt} = u_{p,i} \tag{11.1}$$

$$\frac{Dm_p u_{p,i}}{Dt} = F_{p,i}^{ext} \tag{11.2}$$

$$\frac{Dm_p}{Dt} = \dot{m}_p \tag{11.3}$$

$$\frac{DC_{p,p}T_p}{Dt} = \frac{\Phi_p^c + \Phi_p^{cv}}{m_p}$$
(11.4)

where $C_{p,p}$ is the particle calorific capacity. $F_{p,i}^{ext}$ represents the external forces applied on the particle. \dot{m}_p , Φ^c and Φ^{cv} are the decomposition rate of the particle, the conductive and the convective heat fluxes at its surface respectively. Closure models for \dot{m}_p , Φ^c and Φ^{cv} will be discussed in Chapter 12. In the following, the expression used for $F_{p,i}^{ext}$ is provided.

External forces applied on a rigid spherical particle have been the subject of a number of theoretical works. This canonical problem allows to derive analytical expressions for these various forces. Taking into account the very low ratio between gas and solid densities (ρ_g/ρ_p) of the order of 10^{-3} in all the cases discussed in this thesis), one can reasonably neglect the stationary pressure forces as well as the unsteady "added mass" and Basset force terms in the Basset (B. (1888)), Boussinesq (J. (1903)) and Oseen (Oseen (1927)) equation, so that the only force considered is the drag force. The latter can be expressed, in the case of a flow with uniform pressure, as:

$$F_{D,i} = \frac{1}{2} \rho_g \mathcal{C}_D A \| \mathbf{u}_g - \mathbf{u}_p \| (\mathbf{u}_g - \mathbf{u}_p)$$
(11.5)

where C_D is the drag coefficient and $A = \pi d_p^2/4$ is the projected area of the particle with d_p the diameter of the latter. $\mathbf{u}_g = (u_{g,i})_{i=1,3}$ and $\mathbf{u}_p = (u_{p,i})_{i=1,3}$ are the gas and particle velocities respectively. Closure is obtained once an expression for C_D is provided. The problem is that C_D depends on the flow intensity, which is characterized by the particle Reynolds number:

$$Re_p = \frac{\rho_g d_p \|\mathbf{u}_g - \mathbf{u}_p\|}{\mu_g} \tag{11.6}$$

where μ_g is the gas viscosity. In the simulations presented in Chapter 12, the slip velocity (namely $\|\mathbf{u}_g - \mathbf{u}_p\|$) remain small so that the Reynolds number is in general lower than 5. In this case, Oseen (1927) proposes the following expression for the drag coefficient, taking into account first order inertial effects:

$$\mathcal{C}_D = \frac{24}{Re_p} \left(1 + \frac{3}{16} Re_p \right) \tag{11.7}$$

Therefore, Eq. (11.2) reduces to:

$$\frac{Du_{p,i}}{Dt} = \frac{1}{\tau_p} \left(u_{g,i} - u_{p,i} \right)$$
(11.8)

which introduces a characteristic time $\tau_p = \rho_p d_p^2 / [18\mu_g(1 + (3/16)Re_p)]$. τ_p controls the relax of the particle velocity towards the gas velocity. For very low τ_p , negligible compared to some flow time scale, the particle exhibits a tracer like behavior. For very large τ_p however, the particle is quasi-insensitive to variations of the flow velocity.



Figure 11.1: The solid phase is coupled to the gas phase by introducing additional source terms to the Navier-Stokes equations $\mathcal{S}_{eq}^{p \to g}$. $\mathcal{S}_{eq}^{p \to g}$ at each node takes into account the contribution of all the particles present inside the control volume ΔV surrounding the node.

Coupling with the gas pase

The coupling between the solid and the gas phases is performed by introducing additional source terms to the Navier-Stokes equations in AVBP. Let ΔV be a control volume around a node j of the mesh (see Fig. 11.1) and $N_{\Delta V}$ the number of particles located in this control volume. The source terms for the mass, momentum, energy and mass fractions gaseous conservation equations at the node j:

$$\mathcal{S}_m^{p \to g}(\mathbf{x}_j) = \frac{1}{\Delta V} \sum_{p=1}^{N_{\Delta V}} \Psi_j(\mathbf{x}_p) \dot{m}_p \tag{11.9}$$

$$S_{mom,i=1,3}^{p \to g}(\mathbf{x}_j) = \frac{1}{\Delta V} \sum_{p=1}^{N_{\Delta V}} \Psi_j(\mathbf{x}_p) (-m_p F_{p,i}^{ext} + \dot{m}_p u_{p,i})$$
(11.10)

$$S_{\text{NaOH}}^{p \to g}(\mathbf{x}_j) = \frac{1}{\Delta V} \sum_{p=1}^{N_{\Delta V}} \Psi_j(\mathbf{x}_p) \dot{m}_p$$
(11.11)

$$\mathcal{S}_{E}^{p \to g}(\mathbf{x}_{j}) = \frac{1}{\Delta V} \sum_{p=1}^{N_{\Delta V}} \Psi_{j}(\mathbf{x}_{p}) \left(-m_{p} F_{p,i}^{ext} u_{p,i} + \frac{1}{2} \dot{m}_{p} \|\mathbf{u}_{p,i}\|^{2} - (\Phi_{p}^{c} + \Phi_{p}^{cv}) \right)$$
(11.12)

where $\Psi_j(\mathbf{x}_p)$ is the interpolation function from the particle p to the mesh node j. In AVBP, it is expressed as:

$$\Psi_j(\mathbf{x}_p) = \frac{1/d_j}{\sum_{z=1}^{N_z} 1/d_z}$$
(11.13)

where N_z is the number of edges in the cell (in green in Fig. 11.1).

11.2 Gas-phase inhibitor/flame interaction

This section presents the different steps to derive a mechanism modeling the interaction of the gaseous agent NaOH with the methane/air chemistry, at reasonable cost. This mechanism is composed of two submodels:

• a submechanism for the methane/air chemistry, chosen to be the GRI-mech 3.0 Smith et al. (2017), which is widely used and validated for a large range of operating conditions;

• a submechanism for the interaction of the NaOH gaseous agent with the radical species. Hynes et al. (1984); Williams and Fleming (1999) propose a set of three reactions able to reproduce the first order effect of NaOH on the flame speed:

$$NaOH + H \Longrightarrow H_2O + Na$$
 (R₁)

$$Na + OH + M \Longrightarrow NaOH + M$$
 (R₂)

$$NaOH + OH \Longrightarrow H_2O + NaO$$
 (R₃)

where NaOH depletes the H and OH radical pools.

The resulting full mechanism, referred to as GRI-Inh here, includes 56 species and 328 reactions. However, this model is too expensive to be used in CFD codes, especially in the context of LES. A procedure is therefore needed to reduce the cost of GRI-Inh without altering its prediction capabilities. The YARC reduction tool, developed by P. (2008), is retained in this thesis. It provides a variety of methods to efficiently reduce the computational cost of hydrocarbon mechanisms.

The first step is to identify and remove species that are not relevant to certain operating conditions, which can be done with various techniques. Among the most commonly used methods are the Directed Relation Graph (DRG, Lu and Law (2006)) and the Directed Relation Graph with Error Propagation (DRGEP, Pepiot-Desjardins and Pitsch (2008)) methods, which are based on a graph description of the mechanism. DRGEP is the method available in YARC.

DRGEP approach

The fundamental idea behind this approach is to build a graph representing the interaction between different species in the mechanism. Two species S_i and S_j are connected when they participate in at least one common reaction. In this case, both species are represented by two nodes linked with an edge (see Fig. 11.2). A value $r_{i,j}$ can be assigned to each edge and represents the impact of S_j to the prediction of S_i . In P. (2008), it is expressed as:

$$r_{i,j} = \frac{\left|\sum_{k=1}^{N_{reac}} \nu_{k,i} \dot{r}_k \delta_j^k\right|}{\max(P_i, C_j)}$$
(11.14)

where \dot{r}_k is the rate of the k_{th} reaction, $\nu_{k,i}$ is the net stoichiometric coefficient of species S_i in the k_{th} reaction and δ_i^k is defined by:

$$\delta_j^k = 1$$
 if the k_{th} reaction involves the species S_j (11.15)

$$\delta_i^k = 0$$
 otherwise. (11.16)

 P_i and C_j are used to bound the value $r_{i,j}$ between 0 and 1. Their expression reads:

$$P_{i} = \sum_{k=1}^{N_{reac}} \max(0, \nu_{k,i} \dot{r}_{k})$$
(11.17)

$$C_j = \sum_{k=1}^{N_{reac}} \max(0, -\nu_{k,j} \dot{r}_k)$$
(11.18)

The description of the mechanism by a graph is now complete. Two species S_i and S_j can



Figure 11.2: Part of a Directed Relation Graph (DRG) involving four species. Adapted from P. (2008).

be linked either directly by an edge or via a path involving intermediate species (species A and E in Fig. 11.2 for example). A value $R_{i,j}$ can be assigned to a virtual edge linking two species S_i and S_j that are not directly connected. This is done by taking into account the ensemble \mathcal{P} of paths $p = (S_i, S'_1, S'_2, ..., S'_n, S_j)$ from S_i to S_j , where n < N - 2 and S' any species in the mechanism different from S_i and S_j . A value $r^p_{i,j}$ can then be assigned to each path p:

$$r_{i,j}^p = \prod_{z=1}^n r_{z,z+1} \tag{11.19}$$

 $r_{i,j}^p$ is small for species involved through a long path, in which case species S_i has a very small impact on the species S_i . An expression for $R_{i,j}$ can then be obtained:

$$R_{i,j} = \max_{p \in \mathcal{P}} r_{i,j}^p \tag{11.20}$$

The species with the weakest global interaction coefficients can be removed from the mechanism. An illustration is given in Fig. 11.2. In this example, A strongly interacts with B, and B strongly interacts with E. Because of the product used in Eq. (11.19), the global interaction coefficient $R_{A,E}$ is smaller than $r_{A,B}$ and $r_{B,E}$. This way, direct connections are favored. In this case, if A is the target species that the mechanism must reproduce, removing E is expected to introduce an error which is lower than removing C but higher than removing D.

This method is applied to GRI-Inh leading to the removal of half the species. An additional technique, available in YARC, is used to further reduce the cost of the mechanism. It consists on applying a Quasi-Steady State (QSS) assumption on some species with low concentrations.

QSS assumption

Suppose that a species S_k is in low concentration, with a low net production rate compared to its creation and destruction rates. The conservation equation for the species S_k can be rewritten in terms of concentration c_k :

$$\frac{Dc_k}{Dt} = S_{diff,k} + \dot{\omega}_k^+ - \dot{\omega}_k^- \tag{11.21}$$

where the net production rate $\dot{\omega}_k$ is decomposed into a creation rate $\dot{\omega}_k^+$ and a destruction rate $\dot{\omega}_k^-$. $S_{diff,k}$ represents the contribution of the diffusive flux. When considering

elementary reactions, the destruction term is proportional to the concentration of the species, so that Eq. (11.21) can be rewritten:

$$\frac{Dc_k}{Dt} = S_{diff,k} + \dot{\omega}_k^+ - c_k/\tau_k \tag{11.22}$$

where τ_k is the time scale of species S_k destruction. The QSS assumption holds only if the destruction process is faster that other processes. Thus τ_k is an asymptotically small parameter, so that one can show that $c_k = \tau_k \dot{\omega}_k^+$. For simplicity, only linear coupling is allowed in the present work, and an explicit expression can be derived and integrated in the AVBP solver.

Now that a technique is provided to retrieve the concentration of the QSS species, the only question left is how to find species for which the QSS assumption holds. Several methods can be found in the literature (see Lovs et al. (2000); Lu and Law (2008); Turanyi et al. (1993)). The method used in this work is known as Level Of Importance (LOI) (Lovs et al. (2000, 2002)).

LOI criterion

A steady state parameter can be defined for each species S:

$$Q_S \propto [S]\tau_k \tag{11.23}$$

where [S] is the concentration of the species S and τ_S is a measure of its time scale. In the present case, it is expressed as

$$\tau_k = -\left[\frac{\partial(P_S - C_S)}{\partial[S]}\right]^{-1} \tag{11.24}$$

where P_S and C_S are given by Eq. (11.17,11.18) respectively. The QSS assumption can therefore be applied to species verifying two properties: 1) its concentration remains small; 2) it is highly reactive. The LOI criterion is used to identify the QSS species. The final step of the reduction procedure is to evaluate the QSS species concentrations. Doing so may require to remove some reactions involving two QSS reactants causing quadratic coupling terms. If the removal of these reactions does not weaken the predictability of the mechanism, a linear system is obtained for the QSS species concentrations. An inversion of the system allows to express analytically the concentrations of the QSS species using the other species concentrations.

The whole procedure has been applied to GRI-Inh. The objective is to reduce the mechanism while maintaining a good predictability in terms of laminar flame speed, fuel (CH₄) and gaseous agent (NaOH) concentrations. These three parameters were fixed as targets for the reduction method, which means that the impact of any species removal has been expressed in terms of errors introduced in these targets. The resulting mechanism contains 156 reactions, 20 transported species (i.e. species for which a transport equation is solved) and 8 species in QSS. Among the transported species, radical species like H, OH and O have been kept to ensure that the influence of the gaseous agent NaOH on the flame speed is well reproduced. The main transported species are $CH_4, O_2, H_2O, CO, CO_2, Na, NaOH, H, O, OH...$ and the main QSS species are NaO, CH, HCCO... This mechanism is validated in Chapter 12.

11.3 The TFLES model for multi-step chemistries

The dynamically thickened flame conventionally employed in AVBP was introduced in Sec 3.3.3.3. The theoretical framework at the basis of the TFLES model is still valid in the context of multi-step mechanisms. However, from a more practical point of view, identifying the flame region where the TFLES model must be applied is not straightforward with these mechanisms, as the chemical structure is more complex with hundreds of elementary reaction steps. The unsatisfactory behavior of the standard methodology, when applied to multi-step chemistries, was shown by Jaravel (2016) in his PhD thesis. Jaravel (2016) then redefined a thickening sensor well suited for such mechanisms. It presented in this section.

Jaravel (2016) proposes to base the sensor on the fuel source term $\dot{\omega}_F$:

$$\mathcal{S}_{ARC} = \max\left(\min(2\frac{\mathcal{F}_{max}|\dot{\omega}_F|}{|\dot{\omega}_F|_{1d,max}} - 1, 1), 0\right)$$
(11.25)

where $\dot{\omega}_F$ is computed during the simulation and $|\dot{\omega}_F|_{1d,max}$ is the maximum value for $\dot{\omega}_F$ in a one-dimensional flame corresponding to the operating condition of the simulation. $|\dot{\omega}_F|_{1d,max}$ can be computed by the user using a chemistry solver. It is an input parameter for the simulation. The *min* and *max* operators allows to normalize the sensor between 0 and 1.

The sensor S_{ARC} is able to detect the flame front. It is however too narrow, being localized in the region of strong temperature gradient. To circumvent this problem, Jaravel (2016) proposes to use a scalar Ψ_F , for which a transport equation is solved:

$$\frac{\partial}{\partial t}(\rho\Psi_F) + \frac{\partial}{\partial x}(\rho u\Psi_F) = \nabla (\rho D_{\Psi_F} \nabla\Psi_F) + \rho \dot{\omega}_{\Psi_F}$$
(11.26)

where the source term $\dot{\omega}_{\Psi_F}$ acts as a relaxation towards the sensor \mathcal{S}_{ARC} , allowing Ψ_F to follow it. $\dot{\omega}_{\Psi_F}$ takes the form:

$$\dot{\omega}_{\Psi_F} = \frac{\Psi_F^0 - \Psi_F}{\tau_0} \qquad \text{if } \mathcal{S}_{ARC} > 0.8$$
 (11.27)

$$\dot{\omega}_{\Psi_F} = \frac{0 - \Psi_F}{\tau_1} \qquad \text{if } \mathcal{S}_{ARC} < 0.05 \tag{11.28}$$

When the flame front is detected (i.e. $S_{ARC} \approx 1$), Ψ_F is rapidly relaxed towards the value Ψ_F^0 ; conversely, when S_{ARC} is close to zero, Ψ_F is slowly relaxed towards zero to counteract the diffusion process which tends to broaden the peak of Ψ_F . The final form of the thickening sensor reads:

$$\hat{\mathcal{S}}_{ARC} = \max\left(\min(\Psi_F, 1), \mathcal{S}_{ARC}\right) \tag{11.29}$$

The whole procedure is summarized in Fig 11.3.

The thickening of the sensor S_{ARC} is controlled by 4 parameters: τ_0, τ_1, Ψ_F^0 and D_{Ψ_F} . In practice, τ_0 is chosen of the order of 10 computational time steps. This value is found to preserve numerical stability, while ensuring a rapid relaxation towards Ψ_F^0 . As pointed out by Jaravel (2016), the filtering procedure is weakly dependent on Ψ_F^0 , and a default value has been assigned to it: $\Psi_F^0 = 20$. Finally, D_{Ψ_F} can be evaluated using the smallest



Figure 11.3: Profiles of the quantities used for the sensor \hat{S}_{ARC} . From Jaravel (2016).

Schmidt number of the mixture: $D_{\Psi_F} = \nu/m_k (Sc_k)$. This allows the scalar Ψ_F to be as diffusive as possible, while remaining consistent with diffusion imposed by the species of the chemical scheme. Finally, the only parameter that has to be defined by the user is τ_1 . In practice, τ_1 can be given two values: one for the fresh side and one for the burnt gas side. This procedure has been successfully used on partially premixed swirled burners by Jaravel et al. (2017) and on an autoignition stabilized jet flame by Schulz et al. (2017). Chapter 12

Investigation of the mechanism of flame inhibition by sodium bicarbonate particles

12.1 Influence of sodium bicarbonate powders on selfpropagating flames

Theoretical analysis and simulation of methane/air flame inhibition by sodium bicarbonate particles

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Abstract

The capacity of sodium bicarbonate $(NaHCO_3)_s$ powder to chemically reduce flame speeds and mitigate the effects of accidental explosions is well established. The inhibition of premixed hydrocarbon/air flames by monodisperse $(NaHCO_3)_s$ solid particles is investigated, here, using theory and numerical simulations. First, an analytical solution for the temperature history of a solid $(NaHCO_3)_s$ particle crossing a flame shows that the size of the largest $(NaHCO_3)_s$ particle which can decompose inside the flame front, and act on chemical reactions efficiently, strongly depends on the flame speed. For various fuels and a wide range of equivalence ratios, particles with a strong potential for flame inhibition are identified: hence a criterion, on the maximum particle size, for efficient inhibition is proposed. Thereafter, a one-dimensional methane/air flame traveling in a premixed gas loaded with sodium bicarbonate is simulated using a chemical mechanism based on GRI-Mech, extended to include inhibition chemistry and reduced to 20 species with a DRGEP method [1]. Inhibitor particle size and mass loading are varied to study the flame response to inhibition by (NaHCO₃)_s powders. Finally, two-dimensional simulations of a planar flame traveling in a flow with a non-uniform inhibitor mass loading distribution are analyzed. In the case of strong particle stratification, an acceleration of the flame is observed, instead of a mitigation. This fundamental mechanism may

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limit the actual potential of inhibition powders in real configurations. *Keywords:* Heterogeneous flame inhibition, Sodium bicarbonate, DPS, DNS

1. Introduction

Until the 1970s, industrial safety processes relied heavily on chlorofluorocarbon gases as fire suppressants in ground, sea and air systems [2]. Since then, the threat to Earth's ozone of such gases has been established and their production

- ⁵ prohibited. Following this ban, intensive research has been conducted to find efficient halon replacements. Alkali metal compounds, such as sodium bicarbonate $(NaHCO_3)_s$ have received considerable attention because of their higher effectiveness per mass basis compared to the halon 1301 (CF₃Br) [3]. Extensive experimental investigations on flame inhibition (reduction of flame speed) and
- ¹⁰ flame suppression abilities of $(NaHCO_3)_s$ demonstrate the effectiveness of these powders on premixed flames [4–7] and counter-flow flames [8–11]. More recent laboratory scale and medium scale $(50m^3)$ experiments [12] show the capacity of alkali metal compounds to mitigate the potential aftermath of gaseous explosions.
- ¹⁵ The first systematic investigation of the mechanism of flame inhibition/suppression by (NaHCO₃)_s particles was performed by Rosser et al [4]. The overall mechanism includes two major steps illustrated in Fig. 1: 1) thermal decomposition of sodium bicarbonate particles; 2) gas-phase chemical interaction with the flame.

Particle thermal decomposition. Exposed to a temperature gradient inside the flame front, $(NaHCO_3)_s$ solid particles undergo thermal decomposition. Sridhar Iya et al. [7] demonstrated that particles of $(NaHCO_3)_s$ below $10\mu m$ decompose completely inside the methane/air flame front. Chelliah et al. [11] also showed a monotonic behavior of suppression effectiveness with particle diameter for methane/air flames (contrary to the results of Trees et al. [2] for heptane

flames). These studies indicated that the particle size influences the position at which $(NaHCO_3)_s$ decomposition is complete. Large particles, typically larger than $60\mu m$, will decompose too far downstream of the flame front leading to



Figure 1: Schematic of the flame inhibition mechanism by sodium bicarbonate. Path S-G refers to the path from the Solid bicarbonate particle to the Gaseous agent NaOH. $R\bullet$ stands for a radical species and M denotes a group of stable species.

unsuccessful flame inhibition. The gas temperature decrease during this decomposition phase is generally negligible and does not modify the flame speed significantly.

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Chemical interaction with the flame. The strong potential of sodium bicarbonate powders for flame mitigation is attributed to a chemical rather than thermal (cooling) interaction with the flame. Sridhar Iya et al. [7] found a good correlation between the concentration of Na evaporated from salts and the degree of flame inhibition. They concluded that the mechanism behind flame inhibition by salts is chemical and homogeneous, i.e. taking place in the gas phase as a result of the presence of vaporized inhibitor. More recently Chelliah et al. [11] conducted experiments on the extinction condition of counterflow methane/air flames. Multiple particle size classes were considered. This study demonstrated the dominant chemical nature of flame inhibition by fine (NaHCO₃)_s particles. Since flame inhibition by (NaHCO₃)_s is due primarily to chemical effects, many authors [7, 11, 13–15] have proposed kinetic models based on NaOH being the main gaseous agent, able to act on chemical reactions in the flame. Gaseous sodium hydroxide (NaOH) is formed when the (NaHCO₃)_s particles are exposed to high temperatures inside the flame front. The particles undergo thermal decomposition following a series of reaction pathways (path S-G in Fig. 1) leading to the formation of NaOH. The interaction of NaOH with the flame is homogeneous (gas-phase chemistry) and involves the catalytic recombination of the radical species responsible for the flame consumption speed, namely H, OH and O.

While the main product of the decomposition of the sodium bicarbonate is agreed upon, the path from the $(NaHCO_3)_s$ particle to the gaseous species NaOH (path S-G in Fig. 1) is still uncertain. Hu et al. [16] and Wu et al. [17] proposed a temperature range (370-543K) and kinetic rates for the first stage of the thermal decomposition:

$$(\text{NaHCO}_3)_s \longrightarrow 0.5(\text{Na}_2\text{CO}_3)_s + 0.5CO_2 + 0.5H_2O,$$

However, the following steps lack details and rates. In addition to the uncertainties surrounding $(NaHCO_3)_s$ decomposition kinetics, the influence of the powder injection procedure on the effectiveness of the inhibitor has not been addressed yet. In practice the methods used to deliver the powder to the gas can hardly guarantee a uniform particles distribution. A stratified or segregated powder can perturb the flame front and increase its surface, suggesting a possible opposite effect (increase in flame speed). This behavior was mentioned by Rosser et al. [4]: 'In the absence of powder the flames were approximately hemispherical in shape; ... The presence of powder resulted at times in irregular flame shapes

- ⁴⁰ ... In some cases the flames were very long and highly tilted, probably as a result of non-uniform powder distribution'. More recently, various inhibitors, gaseous and liquid, failed the qualification test performed by the Federal Aviation Administration (FAA). The Aerosol Can Test (ACT) report [18] issued by the FAA showed that in some cases the overpressure increased in presence of
- ⁴⁵ inhibitor agents compared to the case with no agent. Even if the exact cause for these disappointing results is not identified, the powder segregation mechanism may explain it: in the context of vapor cloud explosions (VCE), the inhibitor injection would be performed through a discrete number of high pressure man-

ifolds. Powder segregation is therefore unavoidable and may potentially lead to the amplification of VCE damages. This possibility clearly requires additional studies if such mitigation systems are used in the future.

The present work describes a theoretical and numerical study of the influence of sodium bicarbonate powders on flame propagation. The first objective ⁵⁵ is to derive a model (HetMIS) for the Heterogeneous methane/air inhibition by Sodium bicarbonate suited for numerical simulations in large and complex configurations. HetMIS is composed of: 1) a simplified 1-step model for particle decomposition (Section 2.1); 2) an analytically reduced chemistry (ARC) for flame-NaOH interaction (Section 2.2). The introduction of complex chem-

- ⁶⁰ istry using ARC schemes [19–22] is mandatory to capture the chemical effects of NaOH on the flame at a reasonable cost. An analytical expression for particle temperature is derived in Section 3 to predict where particles decompose in a premixed flame, hence to propose criteria on particle size for complete decomposition inside the flame front. Based on these criteria, a selection of particle sizes
- ⁶⁵ is used to study the effect of sodium bicarbonate powder on the propagation of a 1D flame (Section 4). Finally, the response of a planar flame to a nonuniform distribution of sodium bicarbonate particles is examined in Section 5 using two-dimensional simulations to show that the local flame speed reduction can be offset by a flame surface increase when the distribution of inhibitor is not homogeneous.

2. Model for Heterogeneous methane/air flame Inhibition by Sodium bicarbonate (HetMIS)

The HetMIS model includes a simplified description of the particle evolution from solid $(NaHCO_3)_s$ particles to gaseous NaOH (Section 2.1) and the homogeneous interaction of this gaseous agent with the flame chemistry (Section 2.2).
2.1. Thermal decomposition of sodium bicarbonate particles

 $(NaHCO_3)_s$ first decomposes to form sodium carbonate $(Na_2CO_3)_s$ at temperatures in the range 370-543K [8] depending on the particle size (R_1) . At 1123K [11], the sodium carbonate forms liquid sodium oxide $(Na_2O)_{liq}$ (R_2) :

$$(\text{NaHCO}_3)_{\text{s}} \stackrel{370-543K}{\longrightarrow} 0.5(\text{Na}_2\text{CO}_3)_{\text{s}} + 0.5\text{CO}_2 + 0.5\text{H}_2\text{O} \qquad (R_1)$$

$$(\mathrm{Na}_2\mathrm{CO}_3)_{\mathrm{s}} \xrightarrow{1123K} (\mathrm{Na}_2\mathrm{O})_{\mathrm{liq}} + \mathrm{CO}_2$$
 (R₂)

To produce NaOH, the sodium oxide $(Na_2O)_{liq}$ can follow an heterogeneous reaction (R'_3) or go through phase change followed by an homogeneous reaction (R''_3) .

$$(\mathrm{Na}_2\mathrm{O})_{\mathrm{liq}} + \mathrm{H}_2\mathrm{O} \longrightarrow 2\,\mathrm{NaOH} \quad (R'_3) \quad \left| \begin{array}{c} (\mathrm{Na}_2\mathrm{O})_{\mathrm{liq}} \longrightarrow \mathrm{Na}_2\mathrm{O} \\ \mathrm{Na}_2\mathrm{O} + \mathrm{H}_2\mathrm{O} \longrightarrow 2\,\mathrm{NaOH} \end{array} \right| (R''_3)$$

Major uncertainties surround the thermal decomposition of $(NaHCO_3)_s$ particles. From the formation of sodium carbonate to the production of sodium hydroxide, the only relevant information is the temperature at which $(Na_2CO_3)_s$ decomposes [11] (ie. $T_{decomp} = 1123K$). Therefore, a simplified model is proposed based on a single step approach (R_{glob}) as suggested by Mitani et al. [23, 24] and Chelliah et al. [11]:

$$(NaHCO_3)_s \longrightarrow NaOH + CO_2$$
 (R_{glob})

This reduced decomposition kinetics involves phase change and requires additional equations for the solid particle mass m_p and its temperature T_p . Here, an isolated (NaHCO₃)_s particle, assumed spherical, is described using a Lagrangian formalism. The temperature and density are supposed uniform inside the particle.

NaOH cannot be produced before the solid (NaHCO₃)_s particle has reached T_{decomp} . Therefore, the mass of the particle is kept constant as long as $T_p \leq T_{decomp}$ (Eq. (1)). The only source term left in the particle temperature equation is related to the gaseous conductive heat flux (Φ_q^c):

For $T_p \leq T_{decomp}$:

$$\frac{dm_p}{dt} = 0 \tag{1}$$

$$\frac{dC_{p,p}T_p}{dt} = -\frac{\Phi_g^c}{m_p} \quad \text{where} \quad \Phi_g^c = \pi d_p N_u \frac{\lambda}{C_p} (C_p(T_p)T_p - C_p(T_\infty)T_\infty) \quad (2)$$

where d_p is the particle diameter, T_{∞} is the farfield temperature, λ is the thermal conductivity and C_p the gas calorific capacity. $C_{p,p}$ is the particle calorific capacity. To derive Eq. (2), the term λ/C_p is assumed constant and equal to its value at the particle surface [25, 26]. For all present cases, the slip velocity of the solid particles is small in the fresh gases and increases slowly through the flame so that the particle Reynolds number remains small and the flow around the particle is laminar. Thus, the Nusselt number is $N_u = 2$.

Once T_{decomp} is reached, the particle undergoes fast decomposition (Eq. (3)). During this phase, the temperature of the particle T_p is assumed constant (Eq. (4)). t_p^{\star} is, for each particle, the time when the decomposition temperature is reached: $T_p(t_p^{\star}) = T_{decomp}$. The decomposition duration is fixed by the parameter $\tau_{decomp} = 10\mu s$ and kept small to ensure an almost instantaneous liberation of the gaseous agent.

For $T_p = T_{decomp}$:

$$\frac{dm_p}{dt} = A\rho_p d_p exp(-(t - t_p^{\star})/\tau_{decomp})$$
(3)

$$\frac{dC_{p,p}T_p}{dt} = 0\tag{4}$$

where ρ_p is the particle density and A is a constant set to $1m^2/s$. This model can be summarized in the following equations using the Heaviside function H:

$$\frac{dm_p}{dt} = A\rho_p d_p H(t - t_p^*) exp(-(t - t_p^*)/\tau_{decomp})$$
(5)

$$\frac{dC_{p,p}T_p}{dt} = -\frac{\Phi_g^c}{m_p}H(t_p^\star - t) \tag{6}$$

Such a model is too simple to account for all the details of the inhibition process. In practice, the particles are far from being spherical and can exhibit complex crystalline like structures with varying aspect ratios. Moreover, the model predicts an abrupt stepwise behavior of the inhibitor: if the particle reaches T_{decomp} inside the reaction region, it will decompose completely in the flame zone. Otherwise, it will vaporize without impacting the flame, which is certainly far from being true. Taking into account the temperature distribution inside the particle

- should improve this aspect of the model by reproducing the release of certain amount of Na-containing species inside the reaction zone for large particles due to surface decomposition. However, this should result in a prohibitive increase of the computational cost of the model. The set of equations (5)-(6) are useful to study theoretically and numerically first order effects of sodium bicarbonate particles on flame propagation as long as their decomposition speed is large
 - compared to all other phenomena.

2.2. Kinetic model for homogeneous methane/air flame inhibition

Once NaOH is formed, its interaction with the flame chemistry must be modeled. Hypes et al. [27] and Williams et al. [13], proposed reactions R_{4-6} :

$$NaOH + H \Longrightarrow H_2O + Na$$
 (R₄)

$$Na + OH + M \Longrightarrow NaOH + M$$
 (R₅)

$$NaOH + OH \Longrightarrow H_2O + NaO$$
 (R₆)

where NaOH depletes the H and OH radical pools, explaining why the flame speed is reduced. This set of reactions and the associated species (NaOH, NaO and Na) are incorporated into the GRI-Mech 3.0 mechanism [28] chosen to

⁹⁵ and Na) are incorporated into the GRI-Mech 3.0 mechanism [28] chosen to model methane oxydation chemistry. The resulting full mechanism, referred to as GRI-Inh, includes 56 species and 328 reactions. The YARC reduction tool [29] is then used to derive an Analytically Reduced Chemistry (ARC) with reasonable computational cost. First the DRGEP method [1] is used to eliminate irrelevant species and reactions. Second, a Quasi-Steady State Assumption (QSSA) is applied on highly reactive species. With this approximation, solving a transport equation for the considered species is no longer required: their concentration may be simply computed using analytical expressions. This procedure allows to reduce both the stiffness and the cost of the chemical scheme.



Figure 2: Laminar premixed methane/air flame speeds at atmospheric conditions without inhibitor. (Black line) HomMIS mechanism. (Grey line) GRI-Inh mechanism. Experimental data: (\Box) Dyakov et al. [30]; (\circ) Van Maaren et al. [31]; (+)Vagelopoulos et al. [32]; (\bigtriangledown) Gu et al. [33].

¹⁰⁵ The resulting mechanism contains 156 reactions, 20 transported species (i.e. species for which a transport equation is solved) and 8 species in QSS. It is referred to as HomMIS for Homogeneous methane/air flame Inhibition by Sodium bicarbonate.

The HomMIS mechanism is validated against experimental data, without inhibitor, in terms of laminar flame speed in Fig. 2. The reduced scheme fits the full scheme GRI-Inh within 5% accuracy between $\phi = 0.6$ and $\phi = 1.6$.

A first validation exercise for the inhibition chemistry (R_{4-6}) is to assume that $(NaHCO_3)_s$ particles are small enough to decompose far ahead of the flame front and study the effects of the addition of gaseous NaOH to the premixed

- reactants. A stoichiometric methane/air flame is diluted with variable amounts of NaOH ($Y_{NaOH} = [0..0.02]$) to measure the influence of this gas on the flame velocity, neglecting the solid/gas transformation of the actual inhibition process. The experiments of Rosser et al. [4] can be used as validation data for the homogeneous inhibition mechanism. Indeed, the small size of the particles
- ¹²⁰ used in the Rosser et al. experiments (mean diameter $d_p = 2.3 \mu m$) ensures fast and complete decomposition inside the flame front. The effects of such particles can therefore be attributed to an homogeneous mechanism and compared to



Figure 3: Validation of the HomMIS mechanism on a laminar 1D methane/air premixed flame with gaseous agent NaOH. Left: flame speed. (Solid line) HomMIS mechanism. (\circ) Babushok et al. mechanism [15]. (\Box) Rosser et al. [4]. Right: (Solid line) flame speed. (\Box) H peak mass fraction. (\circ) OH peak mass fraction. (∇) Adiabatic temperature. Each quantity is normalized by its value at zero concentration of NaOH.

the HomMIS model results. Figure 3a shows a good agreement with the experimental data of Rosser et al. [4] and the numerical results of Babushok et al.

- ¹²⁵ [15] in which the kinetic submodel for the interaction between NaOH and the flame is more detailed than the set of reactions R_{4-6} . The flame speed nonlinear response to NaOH is attributed to the rate of consumption of radical species by NaOH (Fig. 3b). For small quantities of the gaseous agent ($Y_{NaOH} \leq 0.005$) a drastic decrease in the radical species peak concentration and consequently in
- ¹³⁰ heat release is observed. However, for NaOH mass faction above 0.005, the flame inhibition process reaches a plateau. This phenomenon is correlated with the radical species concentrations: for $Y_{NaOH} \ge 0.005$, the maximum radical species mass fraction is close to the burnt gas equilibrium value and the scavenging effect of the gaseous agent diminishes. This saturation effect has been addressed
- in details in [34, 35]. Finally, the adiabatic temperature is quasi-insensitive to NaOH concentration. For the small concentrations of NaOH considered here $(Y_{NaOH} \leq 0.02)$, the chemical mechanism of sodium bicarbonate inhibition dominates over the thermal one.



Figure 4: Summary of the paper content.

The HetMIS model described in section 2 is used in the following sections for a phenomenological study of the flame inhibition by sodium bicarbonate particles as summarized in Fig. 4. The particles size distribution is supposed monodisperse in this paper.

3. Influence of $({\rm NaHCO}_3)_{\rm s}$ particle size on the decomposition position

- ¹⁴⁵ Section 2.2 has shown that the chemical description of the gaseous NaOH interaction with flame chemistry was correct when the particles evaporate before entering the flame front. This section focuses on the general case where particles are not infinitely small and proposes an analytical model able to provide the location, relative to the flame front, where the (NaHCO₃)_s particles decompose. Indeed, if the particles are larger than a critical size, they can even cross the flame and decompose downstream of the reaction zone where they will have no effect. The objective of this section is to evaluate the critical diameter d_p^c above which the inhibition effect can not be observed. d_p^c can be defined as the diameter of the largest particle which can decompose inside the region of
- radical species production (δ_R in Fig. 5). d_p^c can be estimated using an analytical solution for the temperature of an isolated (NaHCO₃)_s particle passing



Figure 5: A particle passing through a stationary flame. T_f is the gas temperature. Y_k is a typical radical species mass fraction. χ^* is the position where the particle decomposes. δ_R is the distance from the flame tip (X = 0) to the end of the radical species production region (net production rate $\dot{\omega}_k^{net}$ reaches the adiabatic equilibrium value $\dot{\omega}_{k,ad}^{net}$). δ_R is usually higher than the thermal thickness δ_L^0 and lower than the total flame thickness.

through a premixed stationary flame. Since this section focuses on the particle decomposition position, only the part $T_p < T_{decomp}$ is considered and the chemical interaction with the flame is not treated: the mass and the diameter of the particle are assumed constant.

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3.1. Analytical solution for the temperature of an isolated $(NaHCO_3)_s$ particle crossing a flame

Consider a sodium bicarbonate particle entering a stationary flame (see Fig. 5). The particle is small enough to assume that it is initially at equilibrium with the gas: $u_p^0 = s_L^0$ and $T_p^0 = T_0$ where s_L^0 and T_0 are the laminar flame speed and the fresh gas temperature respectively. The equation for the particle temperature (Eq. (2)) is further simplified assuming constant heat capacity for the particle $C_{p,p}$:

$$\frac{dT_p}{dt} = \frac{\pi \lambda N_u d_p}{m_p C_{p,p}} (T_f - T_p) \tag{7}$$

$$=\frac{6\lambda N_u}{\rho_p d_p^2 C_{p,p}} (T_f - T_p) \tag{8}$$

where T_f is the gas temperature across the flame. Furthermore, the particle velocity is assumed equal to the gas velocity: $u_p = u_f = s_L^0 T_f/T_0$. This assumption is generally too strong to be valid for all particles. However, the particles considered in this section are relatively small ($< 50 \mu m$) and initially at equilibrium with the gas so that this equality is at least true in the preheating zone of the flame. The error on the particle temperature induced by this hypothesis is therefore acceptable as shown in Section 3.2.2. Via variable substitution one can then obtain:

$$\frac{dT_p}{dx} = \frac{1}{u_f} \frac{dT_p}{dt} = \frac{6\lambda N_u}{\rho_p d_p^2 C_{p,p}} \left(1 - \frac{T_p}{T_f}\right) \left(\frac{T_0}{s_L^0}\right) \tag{9}$$

or
$$\frac{dT_p}{dx} = \Gamma_T(d_p, s_L^0) \left(1 - \frac{T_p}{T_f}\right)$$
 (10)

where Γ_T is a temperature gradient coefficient. The gas thermal diffusivity λ is evaluated at $T = T_{decomp}$. To integrate Eq. (10), an expression for the gas temperature T_f is needed. To provide a fully analytical expression for T_p , an analytical expression for T_f is needed. Here, the model of Echekki and Ferziger (EF) [36] is used to express the gas temperature T_f : For $x \leq \delta_{EF}$:

$$T_f = T_0 + (T_{ad} - T_0) \frac{(1 - 1/\beta)}{e - 1} (exp(x/\delta_{EF}) - 1) = C_1 + C_2 exp(\frac{x}{\delta_{EF}})$$

and for $x > \delta_{EF}$:

$$T_f = T_0 + (T_{ad} - T_0)(1 - exp[\frac{(1 - \beta)(x - \delta_{EF})}{\delta_{EF}}]/\beta) = D_1 + D_2 exp[\frac{(1 - \beta)x}{\delta_{EF}}]$$

where β is the nondimensional activation energy for the one-reaction flame model $\beta = T_a(T_{ad} - T_0)/T_{ad}^2$, T_a being the activation temperature. T_{ad} is the flame adiabatic temperature and δ_{EF} is the flame preheat zone thickness defined by $\delta_{EF} = D_{th}/s_L^0$, where D_{th} is the heat diffusivity in the fresh gases.

	$0 \leq x \leq \delta_{EF}$	$x > \delta_{EF}$
$\Phi_T(d_p, s^0_L, T_0, T_{ad}, \delta_{EF})$	$\delta_{EF}\Gamma_T/C_1$	$\delta_{EF}\Gamma_T/[D_1(1-eta)]$
v	$exp(-\frac{x}{\delta_{EF}})$	$exp(-rac{x(1-eta)}{\delta_{EF}})$
G	C_{1}/C_{2}	D_1/D_2
Ŷ	$_{2}F_{1}(1, 1 - \Phi_{T}; 2 - \Phi_{T}; 1 + G)$	$_{2}F_{1}(1,1;1+\Phi_{T};-exp(1-\beta)/G)$
E_T	$T_0^{1-\Phi_T}(1-\kappa\Upsilon)$	$(T_{p,EF} - T_{f,EF}\Upsilon)/(exp(\beta - 1)T_{f,EF})^{\Phi_T}$

Table 1: Particle temperature profile parameters. $T_{p,EF}$ is defined by $T_p(x = \delta_{EF})$, and $T_{f,EF}$ is equal to $T_0 + (T_{ad} - T_0)(1 - 1/\beta)$.

Manipulating this set of equations (substitution and integration by parts), an expression for the particle temperature profile T_p can be deduced (see Annex):

$$T_{p} = \begin{cases} \kappa T_{f\ 2}F_{1}(1, 1 - \Phi_{T}; 2 - \Phi_{T}; 1 + Gv) + E_{T} [vT_{f}]^{\Phi_{T}} & \text{for } x \leq \delta_{EF} \\ T_{f\ 2}F_{1}(1, 1; 1 + \Phi_{T}; -1/[Gv]) + E_{T} [vT_{f}]^{\Phi_{T}} & \text{for } x > \delta_{EF} \end{cases}$$
(11)

The parameters of Eq. (11) are summarized in Table 1. The coefficient κ is equal to $G\left[1-1/(1-\Phi_T)\right]$. $_2F_1(a,b;c;z)$ is the Gauss hypergeometric function. E_T is the integration constant.

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Equation (11) is used to obtain the position χ^* at which the particle will reach T_{decomp} . χ^* can be compared to δ_R , the distance from the flame inlet (X = 0 in Fig. 5) to the end of the region of radical species production. For a given flame (fuel, equivalence ratio, pressure and temperature), δ_R can be evaluated

- using detailed chemical kinetics and deduced from the radical species net production rate profile (see Fig. 5). δ_R is usually higher than the flame thermal thickness $\delta_L^0 = (T_{ad} - T_0)/max(|\frac{dT_f}{dx}|)$ but lower than the flame total thickness defined by the distance from fresh to burned gases. The nondimensional variable $\delta^* = \chi^*/\delta_R$ is therefore used to characterize particle inhibition properties.
- ¹⁷⁵ If $\delta^* < 1$, a reduction of the flame burning velocity is possible since the particle decomposes inside or before the reaction zone; otherwise, NaOH is liberated after the reaction zone, with no interaction with the flame chemistry.

3.2. Validation of the analytical solution

180 3.2.1. Numerical methods

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Section 3.1 gives an analytical expression for the temperature T_p of particles crossing a premixed flame and the position where particles reach the decomposition temperature T_{decomp} . To validate theory, simulations of the configuration considered in Fig. 5 are performed using the DNS/LES solver for the fully compressible multispecies Navier-Stokes equations AVBP, co-developped by CER-

FACS and IFP-EN. A centered continuous Taylor-Galerkin scheme third-order in space and time (TTGC [37]) is used. Gas-phase boundaries are described by Navier-Stokes characteristic boundary conditions [38]. The HomMIS mechanism described in Section 2.2 (Fig. 4) is used to model flame and inhibition the chemistry.

The particles are described using a Lagrangian formalism. The small particle size $(1 - 30\mu m)$ allows to apply the point source approximation. Two-way coupling between solid and gaseous phases is accounted for but collisions are neglected due to the low volume fractions considered here ($\alpha_s < 0.1\%$). Stokes

drag force is applied on the particles. In all simulations, the slip velocity of the particles remains small so that the particle Reynolds number is lower than 5. The drag coefficient then reads $C_d = 24/Re_p(1 + 3/16Re_p)$ [39]. The decomposition of sodium bicarbonate particles is described using the model of Section 2.1.

200 3.2.2. Validation

A sodium bicarbonate particle is placed in front of a stationary methane/air flame in AVBP. The particle is initially at equilibrium with the gas $(u_p^0 = s_L^0$ and $T_p^0 = T_0$).

Figure 6(Left) compares the gas temperature profile across a stoichiometric premixed methane/air flame given by the EF simplified one-step model and by the DNS using the HomMIS mechanism presented in Section 2.2. It shows that the EF analytical model for T_f works reasonably well. Figure 6(Right) shows the particle temperature history extracted from the simulation and compared to the



Figure 6: Left: Stoichiometric laminar premixed methane/air flame at atmospheric conditions using EF simplified one-step model and HomMIS detailed chemistry description. Right: Particle temperature history given by the analytical solution Eq. (11) (dashed lines) is compared to the numerical results obtained with AVBP: $(\nabla) dp = 10\mu m$; (o) $dp = 20\mu m$. T_f is the gas temperature across the flame. The + symbol indicates the position where the particle reaches T_{decomp} according to theory.

analytical solution Eq. (11). A good prediction of the particle decomposition position is observed. For all sizes, particles decompose at locations where the gas temperature is much higher than the decomposition temperature T_{decomp} , showing that the thermal inertia of the particles play a role and that the analytical solution Eq. (11) is useful. The model also confirms that, as the size of the particles increases, they decompose downstream of the flame front as seen in the case $d_p = 20 \mu m$ for example.

Figure 7 displays profiles of gas velocity u_f and particle velocity u_p obtained numerically. Even though the slip velocity is not null and increases with increasing particle size, Fig. 7 shows that the particle velocity remains relatively close to the gas velocity even for large particles ($d_p = 20\mu m$ for example). Therefore,

the assumption $(u_p = u_f)$, used to derive the analytical solution Eq. (11), is acceptable for the particle diameter range considered in this paper. The decomposition position is extracted from the profiles shown in Fig. 6(Right)

for methane/air flames at various equivalence ratios $\phi = 0.8, 1., 1.2$ and con-



Figure 7: Gas velocity u_f and particle velocity u_p history across the flame front obtained with AVBP for two particle diameters: $(\nabla) dp = 10 \mu m$; (o) $dp = 20 \mu m$.

fronted to simulation results in Fig. 8. The influence of the particle diameter d_p on the particle decomposition position is investigated. The range of particle sizes is chosen so that the critical value $\delta^* = 1$ is reached within the limits. A good agreement between theory and simulations is observed. As already suggested by Fig. 6(Right), the position at which the sodium bicarbonate particle decomposes, shifts downstream as the particle size is increased. Beyond 220 $22\mu m$, no inhibition effect is expected for the methane/air flames in the range $\phi = [0.8..1.2]$. The shape of the δ^* vs. d_p curves is attributed to the $1/d_p^2$ dependance of the temperature gradient coefficient Γ_T in Eq. (9).

For a given particle size, the nondimensional decomposition position δ^* is the highest at stoichiometry: $\delta^*(\phi = 1.) > \delta^*(\phi = 1.2) > \delta^*(\phi = 0.8)$. The stoechiometric flame is the most difficult to inhibit for a given particle size. The flame burning velocity follows the same order. This observation suggests a dependance of δ^* on s_L^0 hence u_p^0 since the particle is initially in a thermodynamic equilibrium with the gas. For a given diameter, the particles with higher velocity decompose farther from the initial position explaining the relative position



Figure 8: Influence of particle diameter on the relative decomposition position. Methane/air flames at three equivalence ratios are considered. (solid line) theory. (Symbols) simulations: $(\Box) \phi = 0.8$; (\circ) $\phi = 1$. and $(\nabla) \phi = 1.2$.

of the three curves. Note that δ_R also depends on the equivalence ratio. In this case, $\delta_R(\phi = 1.) < \delta_R(\phi = 1.2) < \delta_R(\phi = 0.8)$, contributing to the result presented in Fig. 8.

An interesting value d_p^c , corresponding to $\delta^* = 1$, can be extracted from Fig. 8. d_p^c is the largest particle diameter that can lead to successful inhibition. This value is computed for various fuels and equivalence ratios in the next section.

3.2.3. Extension of the analysis to other fuels.

Three fuels with increasing burning velocities (methane/air, acetylene/air and hydrogen/air) at various equivalence ratios are considered in Fig. 9. To evaluate d_p^c , the parameters T_{ad} , s_L^0 and δ_R are extracted from 1D laminar premixed flames: the HomMIS mechanism is used for methane/air flames, the Wang et al. [40] detailed mechanism is used for acetylene/air flames and the UCSD [41] detailed mechanism is used for hydrogen/air flames.

The critical powder size for inhibition d_p^c depends strongly on the fuel and on the equivalence ratio. The lowest d_p^c value corresponds to the point of maximum flame speed ($\phi \approx 1.1$ for methane/air flames for example). $d_{p,max}^c \approx 16 \mu m$ is the largest size that can lead to successful inhibition for all atmospheric methane/air flames. Acetylene/air and hydrogen/air flames, due to their higher flame speed and lower thickness, need smaller particles to be inhibited: $d_{p,max}^c$ is reduced to



Figure 9: Critical particle diameter d_p^c (defined by $\delta^*(d_p^c) = 1$) and laminar flame speed s_L^0 for CH₄/air, C₂H₂/air and H₂/air flames at atmospheric conditions and various equivalence ratios. The gray dashed bars [7, 11] and the star symbols [23] represent values extracted from the literature.

 $9\mu m$ and $3\mu m$ for acetylene/air and hydrogen/air flames respectively.

- These results are in good agreement with experimental results found in the literature (Fig. 9). Sridhar et al. [7] studied premixed methane/air flames $(\phi = 1.2)$ inhibited by sodium bicarbonate particles. They concluded that the critical diameter was in the range $[12 - 28]\mu m$ which is coherent with the value $d_p^c \approx 20\mu m$ predicted by the HetMIS model (simulation and theory).
- ²⁶⁵ Chelliah et al. [11], when studying the effect of sodium bicarbonate particles on the extinction conditions of non-premixed counterflow methane/air flame, used three classes of particles. They found that particles in the classes $[0..10]\mu m$ and $[10..20]\mu m$ are effective particles. A dramatic reduction in effectiveness was observed when switching to the class $[20-]\mu m$. They concluded that the critical
- ²⁷⁰ size must be located in the second class which is again coherent with the results given by the model.

An analytical expression for the inhibitor decomposition has also been obtained by Mitani [23]. It predicts that for a hydrogen/air flame at $\phi \approx 0.5$ ($\phi \approx 1.4$ resp.) with a burning velocity of 50 cm/s (300 cm/s resp.), only particles with



Figure 10: Non-dimensional critical particle diameter $d_p^{\star} = d_p^c/\delta_{EF}$ as a function of the equivalence ratio ϕ for various fuels. $\delta_{EF} = D_{th}/s_L^0$ is the flame preheat zone thickness.

size < $17\mu m$ (< $3\mu m$ resp.) will be able to fully decompose in the reaction zone, which is close to the value $d_p^c \approx 13\mu m$ ($4\mu m$ resp.) given by the model. An analytical expression for solid inhibitors decomposition was also derived by Rosser et al. [4], but was applied to NaF and NaCl in the paper. It is worth noting that the analytical expressions of Mitani and Rosser et al. assume a constant particle velocity through the flame front (Eq. (A1) in [23] and Eq. (6) in [4]) contrary to the HetMIS model which accounts for its evolution inside the flame. Moreover, the derivations of Mitani and Rosser et al. do not take into account the relative position of radical species production inside the flame front and its dependence on the fuel (which is done in HetMIS via δ_R). For these reasons, HetMIS should improve the dependance of d_p^c on the combustible (fuel

Eq. (11) also shows that, for a given particle size, T_p depends on the reduced parameter x/δ_{EF} where $\delta_{EF} = D_{th}/s_L^0$. Therefore, it is possible to define a reduced critical particle size for inhibition $d_p^{\star} = d_p^c/\delta_{EF}$. Figure 10 displays d_p^{\star} as a function of the equivalence ratio for the three fuels considered in this

and composition), compared to other approaches found in the literature.

section. It shows that for methane/air flames, the theoretical scaling $d_p^{\star} = 0.27$ is perfect for all equivalence ratios within flammability limits. For acetylene/air flames, the scaling $d_p^{\star} = 0.5$ is also satisfactory. Finally hydrogen, as often, exhibits a specific behavior with a $d_p^{\star} = 0.4$ law valid over a limited range of equivalence ratios but not for very rich flames ($\phi > 1.8$). To first order, however, Fig. 10 confirms that a scaling law based on the reduced critical diameter for inhibition $d_p^{\star} = d_p^c s_L^0 / D_{th} = \eta$ where η is a constant depending only on the fuel is reasonably accurate. Note that, d_p^{\star} may depend on other parameters in practice. Nevertheless, these results can be used to obtain a first order estimation of the range of sizes to use for an efficient sodium bicarbonate powder.

4. Transient behavior of the 1D premixed methane/air flame speed with addition of $(NaHCO_3)_s$ particles.

The previous sections have described how particles decompose in the flame (path S-G in Fig. 1) and how NaOH homogeneous chemical inhibition effect ³⁰⁵ may be modeled. However, the detailed chemical interaction between NaOH and the flame reaction zone at the location of particle decomposition has not been discussed yet. In this section, the flame response to inhibition is studied. Based on the results of the theoretical analysis, a selection of particle diameters around the critical value d_p^c is used to discuss the influence of sodium bicarbonate on the flame consumption speed in Section 4.1. The impact of the inhibitor

Sodium bicarbonate particles are placed in front of a moving 1D premixed methane/air flame at stoichiometry. The cold mixture is at atmospheric conditions. The flame is first initialized without particles to stabilize the flame speed. Then, particles are initialized in the freeh gauge, at the freeh gauge speed, avoid

 $_{\rm 315}$ $\,$ Then, particles are initialized in the fresh gases, at the fresh gas speed, avoid-

mass loading $m_{Inh}^{\star 1}$ is assessed in Section 4.2.

¹ The particles mass loading is $m_{Inh}^{\star} = N_p \rho_p V_p / (m_g + N_p \rho_p V_p)$, where N_p is the total number of particles, V_p is the volume of each particle and m_g is the mass of the gas. The inhibitor mass loading m_{Inh}^{\star} is linked to NaOH mass fraction after total decomposition by: $Y_{NaOH} = W_{(NaHCO_3)_s} m_{Inh}^{\star} / W_{NaOH}$, where W_k denotes the molecular weight of a species k.



Figure 11: A 1D premixed methane/air flame at stoichiometry propagating in a mixture filled with sodium bicarbonate particles. L = 0.1m. The particles are initially in a thermodynamic equilibrium with the gas.

ing any impact of the initialization process on the results. The particles are initially in thermodynamic equilibrium with the gas (Fig. 11). In the following, the consumption speed $(s_{c,het})$ of the propagating flame will be compared to the laminar flame speeds $s_{L,hom}^{\varepsilon}$ shown in Fig. 3a. $s_{L,hom}^{\varepsilon}$ is the laminar speed of a methane/air flame inhibited homogeneously by gaseous NaOH at $Y_{\text{NaOH}} = \varepsilon$. Finally, as mentioned before, radical species reach a peak concentration inside the flame front before being relaxed to their burnt gas equilibrium value. Hence, in the following Y_k^{max} denotes the maximum value reached by a species k inside the flame front (see Fig. 5).

325 4.1. Effect of particle size on flame propagation

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The mass loading of inhibitor introduced is constant and set to $m_{Inh}^{\star} = 0.84\%$. Once fully decomposed, this mass of $(NaHCO_3)_s$ is equivalent to a mass fraction of NaOH: $Y_{NaOH} = 0.004$. Therefore the value $s_{L,hom}^{0.004}$ is used as a reference. The temporal evolution of the flame consumption speed $s_{c,het}$ is presented

- ³³⁰ in Fig. 12. From the results presented in Section 3.2.3, particles with diameter lower than $16\mu m$ should lead to successful inhibition. The present simulations confirm this conclusion: a drastic decay in the flame consumption speed is observed for particles with diameter $d_p = [5, 10, 15]\mu m$. Immediately after the first particles reach the decomposition temperature, $s_{c,het}$ starts decreasing due to
- the interaction between the gaseous agent NaOH and the flame chemistry. $s_{c,het}$ eventually stabilizes at the predicted value $s_{L,hom}^{0.004}$. On the other hand, larger



Figure 12: Temporal evolution of the flame consumption speed $s_{c,het}$ for various inhibitor particle diameters. (Solid line) $d_p = 5\mu m$. (\circ) $d_p = 10\mu m$. (\Box) $d_p = 15\mu m$. (\bigtriangledown) $d_p = 20\mu m$. (x) $d_p = 30\mu m$.

particles $(d_p \ge 20 \mu m)$ decompose downstream of the flame. The latter then remains quasi insensitive to the liberation of NaOH in the burnt gases. Figure 13 displays the effects of the liberated NaOH by particle decomposition on flame

- temperature, heat release and major species profiles for the case $d_p = 10 \mu m$. At the location where (NaHCO₃)_s particles decompose, the production of gaseous NaOH leads to radical species consumption and heat release reduction which causes the flame to slow down. The consumption of NaOH depends on the flame speed and the location of (NaHCO₃)_s decomposition inside the reaction
- zone. Therefore, for the case $d_p = 10 \mu m$, only a fraction of the liberated NaOH finally interacts with the flame reactions. The remaining NaOH is left behind the flame.

As seen in Fig. 12, the flame burning velocity in the case $d_p = 20 \ \mu m$ is not equal to the unhibited flame speed, suggesting a partial effect of the inhibitor in this case. This can be explained by considering NaOH back diffusion. Indeed, even though NaOH is liberated past the reaction zone (at a distance $\Delta = \chi^* - \delta_R > 0$

from the latter), it can diffuse back to reach the exothermic region provided that NaOH mass diffusion velocity (estimated by $s_{NaOH} = D_{NaOH}/\Delta$, with D_{NaOH} NaOH the NaOH mass diffusion coefficient) is greater than the flame



Figure 13: The effect of NaOH liberation inside the reaction zone on: (left) flame temperature T_f (solid lines) and heat release rate H_{RR} (dotted lines); (right) major species profiles for the case $d_p = 10\mu m$. In both figures, lines without symbols refer to the flame prior to inhibition, lines with \bigtriangledown symbol refer to the inhibited flame and lines with \circ symbol represent the particle trajectory. X_f is the position of the leading point of the flame.

- convection speed $(\rho_u/\rho_b)s_L$. In the present case, $\Delta \approx 150 \ \mu m$ and the ratio $(\rho_u/\rho_b)s_L/s_{NaOH} \approx 1.1$ is close to unity. Therefore, a partial reduction of the flame speed can be observed for particle sizes slightly higher than d_p^c .
- Before stabilizing at $s_{L,hom}^{0.004}$, a transient phase can be observed in Fig. 12 for $d_p \leq 15\mu m$. This transient phase is stronger as the size of the particles increases. Indeed, small particles, $d_p \leq 5\mu m$ for example, decompose far ahead of the flame front in a region where the gaseous NaOH efficiently consumes the radical species. Subsequently, $s_{c,het}$ decreases smoothly to reach $s_{L,hom}^{0.004}$. In the case $d_p = [10, 15]\mu m$ however, the particles decompose further away from the flame tip.

365 4.2. Effect of inhibitor mass loading on flame propagation

The particle size is now fixed $(d_p = 5\mu m)$, and the total mass loading of the injected inhibitor is varied $(m_{Inh}^* = [0.21, 0.84, 1.6]\%$ corresponding respectively to $Y_{\text{NaOH}} = [0.001, 0.004, 0.008]$). Figure 14 displays the temporal evolution of the flame consumption speed for the three cases. As expected, after the flame



Figure 14: Temporal evolution of the flame consumption speed $s_{c,het}$ for various inhibitor mass loadings. (o) $m_{Inh}^{\star} = 0.075 mg$. (\Box) $m_{Inh}^{\star} = 0.3 mg$. (\bigtriangledown) $m_{Inh}^{\star} = 1.2 mg$.

crosses the particles path, a drastic decrease in $s_{c,het}$ is observed due to the reduction of the radical species concentrations, thus the heat release. $s_{c,het}$ reaches the expected steady state value corresponding to the inhibitor loading considered. The latter value decreases with increasing inhibitor mass loading.

5. Effect of a non-uniform particle spatial distribution on the propagation of a 2D planar flame

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In the previous sections, the flame inhibition problem has been discussed in its one-dimensional form. Moving to a two-dimensional configuration allows to assess the effect of particles spatial distribution on the flame propagation. In practice, injection manifolds used to disperse inhibition powders in a gas can hardly guarantee an homogeneous particle distribution. Flame speed variations along the flame surface, in the case of inhomogeneous distribution, must be expected and they can induce flame surface increase which may offset the inhibition effects as already observed experimentally by Rosser et al. [4]. Similarly to turbulent flames where the total heat release $\dot{\Omega}$ is the product of the averaged consumption speed $\langle s_c \rangle$ and the flame surface A_f , it is possible that



Figure 15: Particle distribution in the vicinity of a planar methane/air flame at stoichiometry and atmospheric conditions. The channel is 70 mm long and 6 mm wide. Right: Mass density distributions over the vertical axis for the three cases considered. Case 0 corresponds to an homogeneous distribution and Case 2 to the most inhomogeneous one where no particles are present at the top boundary.

a decrease in $\langle s_c \rangle$ due to the inhibitors could be offset by an increase in A_f leading to an overall increase of $\dot{\Omega}$. It is therefore important to investigate possible counter-inhibition effects related to a non-uniform agent distribution.

5.1. Numerical setup

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The 6mm wide and 70mm long 2D channel is illustrated in Fig. 15. A symmetry condition is used for the top and bottom boundaries. A no-slip condition is applied at the left boundary and the right one is an outlet treated with the NSCBC formalism [38]. Initially, the flame is planar and located near the left end of the domain. A methane/air flame at stoichiometry and atmospheric conditions is considered here.

The particles are placed on the right side of the flame front. To ensure a complete decomposition of the particles inside the flame front, the particle diameter is set to $d_p = 5\mu m$ (the critical diameter d_p^c for this case is $16\mu m$). The total mass loading of the injected inhibitor is $m_{Inh}^{\star} \approx 1\%$ corresponding

to $Y_{NaOH} = 0.0047$ and an inhibited flame speed $s_{L,hom}^{0.0047} \approx 0.15 m/s$. A linear stratification is applied in the vertical direction. Three cases are considered. Case 0 corresponds to an homogeneous distribution of $(NaHCO_3)_s$. The inhibitor mass loading gradient is increased for cases 1 and 2. Case 2 corresponds to the most inhomogeneous distribution where no particle is present at the top

405 boundary.

5.2. Results

The flame shape at different times is presented in Fig. 16. For the homogeneous case (case 0), the flame remains planar and the flame slows down similarly to the 1D simulations in section 4. The resulting displacement speed is approximately 1.1m/s and is close to the predicted value $T_{ad}s_{L,hom}^{0.0047}/T_0$. As expected, an increase of flame surface is observed for cases 1 and 2 as a result of the inhomogeneous distribution of (NaHCO₃)_s particles. The finger shape exhibited by the flame in cases 1 and 2 is due to the strong local flame speed variations along the flame surface, which creates a strong flame stretch and leads, after 5ms, to a global acceleration of the flame.

The remarkable difference in total flame lengths between cases 1 and 2 is attributed to the flame stretch caused by the variations of the local flame speed along the flame surface. These variations are due to the non-linear response of the flame speed to NaOH concentration associated with the "ideal inhibitor

⁴²⁰ model" (Fig. 3a). Figure 17 displays the NaOH mass fraction along the flame surface averaged over the progress variable c and the distribution of the local flame consumption speed s_c at t = 2.5 ms. Theses curves change with time and are plotted in Fig. 17 when the flame starts interacting with particles. As expected, the local consumption speeds are all smaller than the uninhibited value

 $s_{L}^{0} \approx 0.39 m/s$ but they are also changing strongly from bottom to top for cases 1 and 2 explaining the flame surface increase observed in Fig. 16. Figure 18 displays the temporal evolution of the flame surface A_{f} and of the mean heat release $\dot{\Omega}$ in the channel. The flame response to an homogeneous

particle field (case 0) is as expected: the flame remains planar, its surface is constant $(A_f/A_f^0 = 1)$ and the mean heat release in the chamber drops significantly as a result of inhibition. For a stratified distribution however, the curve exhibits a parabolic shape. For case 2, the inhibition effect is dominant before $\approx 4.2ms$ and a negative slope is observed in the $\dot{\Omega}$ curve. However, since the inhibition effect is not homogeneous along the flame front, the flame area



Figure 16: Propagation of a 2D initially planar methane/air flame in a inhomogeneous distribution of $(NaHCO_3)_s$ particles. The flame shape (isosurface of progress variable c) and the corresponding time in milliseconds are shown for cases 0, 1 and 2.



Figure 17: Inhomogeneous flame displacement speed along the flame surface as a result of stratified particle distribution (cases 0, 1 and 2) at t = 2.5 ms. Left: NaOH mass fraction along the flame surface averaged over the progress variable c. Right: Flame consumption speed along the flame surface.

- increases. At $t \approx 4.2ms$ (marked by *), the flame surface effect dominates the local flame speed reduction so that the total heat release starts increasing. By the time the flame reaches the end of the domain, the heat released inside the chamber has more than doubled as a result of the flame surface increase. For both cases 1 and 2, $\dot{\Omega}$ at the end of the simulation exceeds its initial value.
- Figure 18 underlines possible counter-effects to flame inhibition related to nonuniform particle distributions. This phenomenon might also explain overpressure increase observed in the FAA-ACT experiments [18] where the introduction of inhibition powders actually led to faster flames and higher overpressures. Many authors [42–45] have proposed an explanation for the observed overpres-
- ⁴⁴⁵ sure increase based on thermodynamic equilibrium calculations and perfectly stirred reactor simulations. A non-uniform inhibitor distribution prior to ignition, and the subsequent flame surface increase, may also explain the increase in pressure during these experiments. This is particularly true since agent distribution was not characterized in the aerosol can test experiments. In real



Figure 18: Flame response to $(NaHCO_3)_s$ particles distribution. Left: temporal evolution of the flame surface normalized by its initial value. Right: Temporal evolution of the mean heat release in the channel. The symbol * indicates the slope change in the heat release evolution for case 2 related to the transition from planar to finger shaped flame.

explosions, the presence of obstacles in the path of a flame might add up to the above effects and lead to damages amplification instead of the desired mitigation of explosion hazards.

6. Conclusion

Inhibition of hydrocarbon/air flames by monodisperse solid sodium bicar-⁴⁵⁵ bonate particles has been studied numerically and analytically. The higher effectiveness per mass basis of sodium bicarbonate compared to other inhibitors motivated the choice of (NaHCO₃)_s. This efficiency is attributed to the chemical nature of flame/(NaHCO₃)_s interaction. (NaHCO₃)_s particles undergo thermal decomposition when exposed to flame temperature. As a result, a gaseous agent ⁴⁶⁰ NaOH is liberated and acts as radical species scavenger reducing heat release,

NaOH is liberated and acts as radical species scavenger reducing heat release, hence flame speed.

To account for the path from $(NaHCO_3)_s$ to NaOH, a simplified thermal decomposition model is proposed based on a single step approach. Although it is too simple to account for the details of particle heating and decomposition, this

465 model is able to reproduce first order effects. The diameter of the largest par-

ticle d_p^c able to decompose inside the flame front for different fuels is evaluated using an analytical expression of particle temperature and is consistent with the literature. These results show that, unless the samples are finely crushed, using the same powder without a clear understanding of its limits may lead

- to unsuccessful inhibition in the case of highly reactive fuels for instance. For safety reasons, the particle size distribution inside the inhibitor injection tanks must be carefully characterized. A simple scaling law giving the maximum particle size d_p^c in a sodium bicarbonate powder used to inhibit a given flame is proposed from theory and simulations and reads $d_p^c s_L^0/D_{th} = \eta$, where η is a
- constant depending only on the fuel: η = 0.27 for methane/air flames, η = 0.5 for acetylene/air flames and η = 0.4 for hydrogen flames.
 To reproduce NaOH interaction with the gas phase chemical reactions, an Analytically Reduced Chemistry (ARC) is derived and validated against both Rosser
- et al [4] experimental data and Babushok et al. [15] detailed chemistry findings. This mechanism is coupled with the single step decomposition model to investigate the impact of sodium bicarbonate particles on the propagation of a 1D stoichiometric methane/air flame. This allows to describe transient effects of flame inhibition. Eventually, the flame consumption speed reaches a steady state value dependent on the mass of injected inhibitor: this speed is coherent
- ⁴⁸⁵ with the homogeneous inhibition results. Finally, the effect of spatial inhibitor distribution on the propagation of a 2D stoichiometric methane/air flame is discussed. This simulation highlights possible counter-inhibition effects: as a response to sodium bicarbonate non-uniform distribution, the flame is highly stretched leading to a global flame accelera-
- tion instead of mitigation. This result echoes experimental observations where the introduction of inhibition powders actually increased the overall combustion speed and the overpressure [4, 18].

In terms of applications, to lead to an efficient inhibition, two conditions must be met: (1) the powder size must be smaller that a limit value d_p^c which de-

⁴⁹⁵ pends on the flame thickness and speed (because of their higher flame speed and lower flame thickness, hydrogen/air flames require much smaller particles than methane/air flames for example); (2) the inhibitor distribution in the mixture must be quasi-homogeneous, otherwise flame acceleration can be observed. Property (1) is easy to ensure and the proposed simple scaling law $d_p^c = \eta D_{th}/s_L^0$

can be used to set up an efficient powder. Property (2), however, will depend on the powder injection system and is much more difficult to achieve. From the fire safety point of view, the side effects, related to powder stratification, may be easily circumvented ensuring the delivery of a minimum mass needed to suppress a given flame. However, from the perspective of gas explosions, the objective is to avoid extinguishing the flame since after flame suppression, the volume remains filled with a reactive mixture susceptible to ignite and to

explode. It is then of crucial importance to verify property (2).

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Annex

The equation for particle temperature Eq. (10) reads:

$$\frac{dT_p}{dx} = \Gamma_T(d_p, s_L^0) \left(1 - \frac{T_p}{T_f}\right)$$

After integration, the particle temperature equation becomes:

$$Tp = \frac{\Gamma_T \int \nu(x) dx + C}{\nu(x)} \tag{12}$$

where $\nu(x) = exp \left(\Gamma_T \int [1/T_f] dx \right)$.

The expression of the flame temperature T_f , given by (EF) model, depends on the position relative to δ_{EF} . However, the nature of T_f expression for $x \leq \delta_{EF}$ is identical to that of T_f for $x \geq \delta_{EF}$. Therefore, in the following, the derivation of the analytical expression Eq. (11) is detailed for the case $x \leq \delta_{EF}$ only. For $x \leq \delta_{EF}$:

$$\begin{split} \int \frac{1}{T_f} dx &= \int \frac{1}{C_1 + C_2 exp(\frac{x}{\delta_{EF}})} dx \\ &= \frac{x}{C_1} - \frac{\delta_{EF}}{C_1} ln \left(C_1 + C_2 exp(\frac{x}{\delta_{EF}}) \right) \end{split}$$

Therefore,

$$\nu(x) = \left[C_1 exp(-\frac{x}{\delta_{EF}}) + C_2\right]^{-\Gamma_T \delta_{EF}/C_1}$$
(13)

The only term left in Eq. (12) is,

$$\int \nu(x)dx = -\delta_{EF} \int \frac{\left[C_1 v + C_2\right]^{-\Gamma_T \delta_{EF}/C_1}}{v} dv \text{ with } v = \exp(-\frac{x}{\delta_{EF}})$$
(14)

$$= -\delta_{EF} C_2^{-\Gamma_T \delta_{EF}/C_1} \int \frac{g^{-\Gamma_T \delta_{EF}/C_1}}{g-1} dg \text{ with } g = 1 + \frac{C_1}{C_2} v \qquad (15)$$

Here,

$$\begin{split} g &= 1 + \frac{C_1}{C_2} v \\ g &= 1 + \left(\frac{T_0(e-1)}{(T_{ad} - T_0)(1 - 1/\beta)} - 1 \right) v \\ g &= 1 + \left(\frac{e-1}{(\sigma - 1)(1 - 1/\beta)} - 1 \right) v \text{ with } \sigma = T_{ad}/T_0 \end{split}$$

Since in typical flames $\sigma > 3$. and $\beta > 2$, one can prove that:

$$\forall x \in]0, \delta_{EF}], |g| < 1$$

Therefore, the integral 15 is absolutely convergent. Using Taylor series expansions, one can link this integral to the gauss hypergeometric function :

$$\int \frac{g^{-\Gamma_T \delta_{EF}/C_1}}{g-1} dg = -\int \frac{g^{\Omega}}{1-g} dg \qquad \text{with } \Omega = -\Gamma_T \delta_{EF}/C_1$$
$$= -\int g^{\Omega} \sum_j g^j dg$$
$$= -\sum_j \frac{g^{j+\Omega+1}}{j+\Omega+1}$$
$$= -\frac{g^{\Omega+1}}{\Omega+1} \sum_j \frac{\Omega+1}{j+\Omega+1} g^j$$
$$= -\frac{g^{\Omega+1}}{\Omega+1} \sum_j \frac{(1)_j (\Omega+1)_j}{(\Omega+2)_j} g^j / j!$$
$$= -\frac{g^{\Omega+1}}{\Omega+1} {}_2 F_1(1, 1+\Omega; 2+\Omega; g) \qquad (16)$$

 $(a)_n$ is the Pochhammer symbol defined as :

$$(a)_0 = 1,$$
 $\forall n > 0, (a)_n = \prod_{k=1}^n (a+k-1)$

The Gauss Hypergeometric function is defined for any $a \in \mathbb{C}$, $b \in \mathbb{C}$ and $c \in \mathbb{C} \setminus \{\mathbb{Z}^- \cup \{0\}\}$ by the following Taylor-Series expansion:

$$_{2}F_{1}(a,b;c;z) = \sum_{j=0}^{\infty} \frac{(a)_{j}(b)_{j}}{(c)_{j}} \frac{z^{j}}{j!} \text{ if } |z| < 1$$

Combining Eq. (13) and Eq. (16) with Eq. (12) leads to the analytical expression Eq. (11).

For the case $x > \delta_{EF}$, the steps are very similar. The integral in Eq. (14) becomes:

$$\int \nu(x)dx = -\frac{\delta_{EF}}{1-\beta} \int \frac{[D_1v' + D_2]^{-\Gamma_T\delta_{EF}/((1-\beta)D_1)}}{v'}dv'$$
(17)

where $v' = exp(-\frac{(1-\beta)x}{\delta_{EF}})$. To end up with a convergent integral, the following variable substitution is proposed:

$$g' = \frac{D_2}{D_1 v'}$$
 with $\forall x \ge \delta_{EF}$, $|g'| \le \frac{\sigma - 1}{\beta \sigma} < 1/\beta$

One can still retrieve the Gauss hypergeometric function, only with different parameters (Eq. (11)).

Finally, the Gauss hypergeometric series converges for |z| < 1 but the number of terms needed to reach an accurate value depend on the value of |z|. This number is significantly reduced if |z| < 1/2. Therefore, one is advised to use the final transformation formulae after the step 16:

$$g' = \begin{cases} g & \text{if } 0 \le |g| \le 1/2 \\ 1 - g & \text{if } 1/2 < |g| < 1 \end{cases}$$

Chapter 12. Investigation of the mechanism of flame inhibition by sodium bicarbonate particles

12.2 Annexe: Deflagration inhibition by sodium bicarbonate: a demonstration case

Section 12.1 presented a numerical and theoretical investigation of the impact of sodium bicarbonate particles on self-propagating one-dimensional and two-dimensional flames. Their effect on gas explosions, where turbulence is involved, still needs to be explored. In the absence of well documented experimental studies of explosion mitigation by metal salts, it has been decided to use the Masri et al. (2012) small scale explosion chamber to perform a demonstration case. Indeed, this configuration has already been extensively studied using LES and in particular using AVBP by Quillatre et al. (2013); Volpiani et al. (2017a); Vermorel et al. (2017). It is well suited for LES and model validation since its dimensions are small and its initial and boundary conditions are perfectly defined. The objective here is to demonstrate the suitability of the decomposition model for 3D LES of gas explosions.

12.2.1 Presentation of the BBBS case

Masri et al. (2012) proposes a small scale vented explosion chamber to study the response of various flames on repeated obstructions. It is composed of a square cross section channel $(50 \times 50 \times 250 \text{ }mm^3)$ with solid obstacles. The obstacles can be placed following various arrangements, but only one of them (called BBBS) is retained for the present study. The channel is detailed in Fig. 12.1. The left side of the channel is closed, whereas the right side is opened out to the atmosphere. Three series of obstacles are placed 20 mm, 50 mm and 80 mm away from the ignition source, located at the center of the left closed end of the channel. Each obstruction consists of 5 strips, 4 mm wide, equally separated by 5 mm gaps. An additional obstacle, $12 \times 12 \text{ }mm^2$ cross section, is placed downstream of the baffle plates 94 mm away from the ignition point.

Among all available fuels, a focus is made on the compressed natural gas (CNG: 88.8% CH₄). The fuel/air mixture is initially at rest and ignited by focusing a laser at the closed end of the chamber. The experimental database includes pressure-time measurements and flame front visualizations based on high-speed laser induced fluorescence OH (LIF-OH) imaging.

Numerical setup

The numerical setup is similar to the one used by Quillatre et al. (2013); Vermorel et al. (2017). The three-dimensional computational domain comprises the combustion chamber and a plenum attached to its open end. This plenum mimics the atmosphere and avoids imposing a boundary condition at the exit of the chamber. It also allows a better handling of acoustic waves, especially after the peak pressure induced by the accelerating flame. The mesh size in the chamber is uniform and equal to 0.34 mm. In the plenum, the grid is gradually coarsened. The mesh contains about 60 M cells. Simulations are performed with the TTGC convection scheme. The walls of the chamber and obstacles are modeled using isothermal law of the wall. For the atmospheric outlet, the NSCBC formalism is prescribed. The WALE model describes the subgrid stress tensor. The explosion is ini-



Figure 12.1: Top view of the studied combustion chamber (Masri et al. (2012)). Dimensions are in millimeters. The pressure transducer is located in the center of the closed end and is represented by the red star symbol

tialized by an hemi-spherical flame of radius 5 mm. A one-dimensional laminar flame profile at the operating conditions is used for the transition from fresh to burnt gases. For the solid phase (sodium bicarbonate), the Lagrangian formalism described in Section 11.1 is used. The decomposition model is the one detailed in the first part of this chapter.

The CNG fuel is replaced by a stoichiometric methane/air mixture as in Quillatre et al. (2013). The HomMIS model is used for the flame chemistry. The mesh resolution does not allow to correctly resolve the flame front, so that the TFLES model is used to thicken the flame. The thickening sensor is the one described in Section 11.3, which is well suited for multi-step chemistries. The model of Colin et al. (2000) is used.

Sodium bicarbonate particles are introduced at rest in the fresh gases before the start of the simulation. The particles are uniform in size, and placed homogeneously in the channel. The size of the particles is $d_p = 10\mu m$, which guarantees a complete decomposition of the particles inside the reaction zone of the flame. The only parameter varied is the total mass of inhibitor. Three mass loadings are considered: $m_{Inh}^{\star} = [0, 0.63, 1.26]\%$. They correspond to a mass fraction of NaOH after decomposition $Y_{NaOH} = [0, 0.003, 0.006]$, in accordance with the values used in the paper.

Results

Figure 12.2(Left) displays the evolution of the flame tip velocity s_{tip} as a function of the flame tip position X_{tip} for increasing inhibitor mass loadings. When no particle is injected in the chamber, the flame undergoes repeated accelerations due to flame/obstacle and flame/turbulence interactions. The evolution of s_{tip} is in good agreement with the LES results of Quillatre et al. (2013) performed with a two-step chemistry. Differences appear between both curves between obstacle 3 and 4, which is attributed to the increased resolution around the obstacles adopted in our simulations.

When particles are introduced, the acceleration of the flame is less pronounced due to the inhibitor induced decrease of the flame consumption speed. The flame tip velocity is normalized in Fig. 12.2(Right) by the laminar speed s_L^{ε} of a methane/air flame diluted with $\varepsilon = [0, 0.003, 0.006]$ mass fraction of NaOH. Consider cases $m_{Inh}^{\star} = [0, 0.63]\%$
Chapter 12. Investigation of the mechanism of flame inhibition by sodium bicarbonate particles



Figure 12.2: Flame tip velocity s_{tip} (Left) and non-dimensional flame tip velocity $s_{tip}/s_L^{\varepsilon}$ (Right) as a fonction of the flame tip position X_{tip} for increasing sodium bicarbonate mass loadings m_{Inh}^{\star} . s_L^{ε} is the laminar flame speed of a stoechiometric methane/air flame diluted with a mass fraction ε of NaOH. The LES results of Quillatre et al. (2013) are displayed in red dots.

first. The collapse of the non-dimensional flame tip velocity curves indicates that s_L^{ε} is the controlling parameter. In the last part of the channel, a deviation of the particle laden curve from the particle-free curve is observed, which is attributed to lower turbulent fluctuations ahead of the flame in the former case due to the less intense flame acceleration. The heavy laden case, however, seems to exhibit a singular behavior. While the evolution of the non-dimensional flame speed prior to obstacle 3 is similar to the previous cases, a clear separation appears when the flame exits obstacle 3. This can be explained by the very low flame propagation speed in the heavy laden case: it is almost three (five resp.) times lower than the case $m_{Inh}^* = 0.63\%$ ($m_{Inh}^* = 0\%$ resp.) when the flame exits the third obstruction. This leads to very low turbulent fluctuations ahead of the propagating flame compared to the other cases. The lost contribution of the flame wrinkling is primarily responsible for this singular behavior.

Figure 12.3(Left) shows the temporal evolution of the overpressure in the channel at a probe located at the center of the ignition plate (red star in Fig. 12.1). The consumption of radical species by the gaseous agent decreases the overall heat release rate of the flame, which leads to a drastic decrease of the peak overpressure. This suggests that sodium bicarbonate powders can be used efficiently to mitigate gas explosions when injected homogeneously in the chamber. Both pressure and time are normalized in Fig. 12.3(Right) by s_L/R and $\rho_u(s_L^{\varepsilon})^2$ respectively. The non-dimensional peak overpressures, observed in the cases $m_{Inh}^* = 0\%$ and $m_{Inh}^* = 0.63\%$, are vey close, which suggests that the reduction of the maximum overpressure can be deduced from the ratio of laminar flame speeds squared: $(s_L^{\varepsilon}/s_L)^2$. The non-dimensional overpressure peak observed in the heavy laden case is lower than in the other cases, which can be attributed to the lost contribution of turbulence to the flame surface.

Finally, it is important to note that in these three cases, no flame suppression occurred. A test case with a larger quantity of inhibitors ($\varepsilon = 0.015$) has been computed, in which the heat losses as well as the drastic depletion of the radicals led to flame suppression.



Figure 12.3: Left: Temporal evolution of the overpressure at a probe located at the center of the ignition plate (red star in Fig. 12.1) for increasing sodium bicarbonate mass loadings m_{Inh}^{\star} . s_L^{ε} is the laminar flame speed of a stoechiometric methane/air flame diluted with a mass fraction ε of NaOH. Right: Both time and overpressure are normalized by s_L/R and $\rho_u(s_L^{\varepsilon})^2$ respectively. R is the radius of the channel.

Conclusions

This thesis presents a numerical study of gas explosion phenomena in confined and semiconfined configurations. It focuses on explosions that are initiated by a mild ignition leading to the formation of a subsonic flame. When the latter propagates in an environment with high levels of confinement and congestion, which is the case in most industrial sites, a Flame Acceleration (FA) process is often observed that can give rise to very fast flames, known for their destructive potential. The outcome of the explosion scenario depends on various parameters. Three specific scenarios are covered in this thesis:

- No mitigative procedure available: in this case, the self-propagating flame can freely accelerate. A numerical study is proposed to understand the flame response to repeated obstructions and strong confinement. This investigation is based on the Gravent explosion channel, setup at the university of Munich TUM. In accordance with the experimental results, the numerical simulations highlighted a strong influence of the mixture composition on the explosion scenario:
 - for a lean premixed hydrogen/air mixture, a strong flame acceleration is observed experimentally. The characteristic features of the explosion are well reproduced numerically using a Large Eddy Simulation (LES) approach. The role of confinement as well as the crucial importance of repeated flame-obstacle interactions in producing very fast deflagrations is highlighted. Obstacles impact the flame propagation via two main ingredients: 1) the flow contraction accelerates the flow ahead of the flame and increases the burning velocity of the latter; 2) vortex shedding, visible downstream of the obstacles, wrinkles the flame and in turn increases its burning velocity. The combination of both ingredients can give rise to very fast flames, supersonic relative to a fixed point, and capable of producing strong shocks even without transition to detonation.
 - for a stoichiometric hydrogen/air mixture, a much stronger acceleration phase is observed, which creates the appropriate conditions for DDT. A focus is made on the instants surrounding the DDT event, using Direct Numerical Simulations (DNS). Particular attention is drawn to the impact of the chemistry modeling on DDT. A single-step mechanism, commonly used in DDT simulations, is confronted to a detailed mechanism. The numerical simulations show that the DDT mechanism itself is strongly impacted by the chemistry description. It is shown that the spontaneous initiation of detonations can be triggered, in the case of single-step mechanisms, for conditions of temperature far less restrictive than the ones predicted by multi-step chemistries, including

conditions that can be considered frozen (i.e. for which the time scale of ignition is higher that the time scale of flame propagation). This raises questions about the suitability of single-step chemistries for DDT simulations. Taking into account the time scale of flame propagation, it is shown that DDT, via spontaneous initiation and in the case of detailed chemistries, requires strong shocks able to bring the mixture locally to high temperatures, which can be very difficult to achieve. It is shown that shocks can induce DDT via another mechanism by interacting with the flame internal structure. A pressure pulse amplification can then occur inside the flame brush. The presence of highly reactive species in detailed chemistries is shown to facilitate this mechanism.

• Inhibiting powders can be injected ahead of the flame: well-controlled experiments showed the high efficiency of metal salts (like sodium bicarbonate) in inhibiting fires, which motivated a potential use for gas explosion safety. However, their applicability to the inhibition of self-propagating flames is not straightforward and requires additional experimental and numerical work. The last part of the thesis focuses on the mechanism of transient flame inhibition by sodium bicarbonate particles. First, criteria based on the particle sizes are established to characterize the inhibition efficiency of the particles. Second, two dimensional numerical simulations of a planar flame propagating in a stratified layer of very fine sodium bicarbonate particles showed that under certain conditions these powders can act as combustion enhancers. These results echo a number of experimental observations on the possible counter-effects of the inhibitors.

Perspectives

The different points discussed in this thesis, open the way to further investigations:

- FA:
 - A single configuration has been considered. The influence of both the blockage ratio and the obstacle spacing has not been assessed, despite their crucial importance. In the Gravent database, both parameters were varied. Numerical simulations of these additional configurations should improve our understanding of the flame/obstacle interaction.
 - the efficiency function used in this thesis was based on a constant β fixed by the user. It would be interesting to test dynamic formulations of β , as was done by Volpiani et al. (2017a), especially since the gap between the obstructions is large and allows a re-laminarization of the flow. A dynamic approach is clearly more appropriate in this case.
- DDT:
 - the DDT simulations highlighted the strong impact of the chemistry on the DDT mechanism. Further investigations are needed to fully comprehend this influence. The main problem is the complexity of the scenario and the nonuniqueness of the process that can trigger it. This makes it difficult to identify the essential physics on which simplified mechanisms can be fitted on. One

Chapter 12. Investigation of the mechanism of flame inhibition by sodium bicarbonate particles

could conclude that the way to go is to use detailed chemistries. However, many investigators argue that the conditions under which DDT occurs are far from the validity span of these mechanisms, which adds complexity to the problem. For this reason, a parametric study of the phenomenon in a much simpler configuration has to be conducted. For instance, a one-dimensional computation of the interaction of a fast flame with a counter-advancing strong shock can be performed with various mechanisms. The requirements in terms of flame speed and shock Mach number for DDT triggering can then be characterized. This should lead to a fine description of the pressure pulse amplification process.

- the DDT simulations highlighted our difficulty to properly reproduce the internal structure of detonation fronts in the case of detailed chemistry. Numerical simulations of the propagation of a one-dimensional detonation are needed to correctly address this issue. Once a suited methodology for detailed mechanisms is obtained, the next step is to perform a sensitivity analysis of the detonation structure to chemical reactions.
- Inhibitors: the flame inhibition by sodium bicarbonate was investigated here using very simple configurations, far from representing the key features of gas explosions. Parameters such as turbulence, particle size distribution and injection systems were not discussed in this thesis. The main problem is the lack of well documented gas explosion experiments with sodium bicarbonate powders. Small scale experiments, on which numerical simulations can be validated, are of vital importance towards both the understanding of the phenomena and assessing the applicability of the powders on gas explosions. This is the objective of an ongoing collaboration with TOTAL company.

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