Modelling of Propagating Spherical and Cylindrical Premixed Flames

PROEFSCHRIFT

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Preface

Since ancient times our main source of energy has been the combustion of fossil fuels, and despite efforts to convert to alternatives this situation is unlikely to change in the near future. Throughout centuries, combustion processes have been proven to be a flexible energy source that can be applied in, for instance, domestic heating, electrical appliances, and transportation. The main disadvantages of burning fossil fuels in this manner are the finiteness of our natural resources, and the corresponding emission of pollutants such as nitrogen oxides, soot, and unburnt hydrocarbons. This might have a devastating effect on our ecological environment. As a possible solution to these problems, different kinds of energy production have been developed, using e.g. wind, solar, and nuclear energy sources. At present however, more than eighty percent of our daily energy requirements are still fulfilled by the traditional combustion of fossil fuels. It is therefore important to make these combustion processes as efficient as possible. Until a few decades ago, research on this topic was mainly empirical. However, to fulfill the current, very restrictive governmental regulations, more detailed knowledge of the fundamental processes is required. Besides reducing pollutant emissions and optimizing machinery, this knowledge is also very useful to decrease safety hazards, and of course to simply satisfy our scientific curiosity.

Combustion can have different forms: combustion of gases (e.g. natural gas and biogas), liquids (e.g. petrol and kerosine), and solids (e.g. wood and coal). These combustion processes can generate premixed or non-premixed flames. In premixed flames, the fuel and oxidizer are mixed before the mixture is ignited, for example in a central-heating boiler. In non-premixed flames, the fuel and oxidizer are separated, for example in diesel engines, where the fuel spray is injected into the compressed air in the cylinder. In these flames, the fuel mixes with the oxidizer due to convection and diffusion processes in the flame itself. It is also possible for flames to contain both premixed and non-premixed parts, for example in triple flames. These flames are called partially premixed flames. In this thesis, only premixed flames are studied. The gas mixture consists of methane and air with an unburnt temperature of 300 K. Furthermore, only flames under atmospheric pressure are considered. Both laminar and turbulent flames are investigated. The laminar flames are perfectly spherical or cylindrical, which can either expand, implode, or remain steady. The considered turbulent flames are expanding flames in a turbulent flow field.
The interest in laminar spherical and cylindrical flames is widely spread, because these flames are very useful for determining crucial parameters in premixed combustion such as the burning velocity, flame stretch rate and flame curvature. Extended numerical and experimental research in this area has been performed in numerous studies. Experimental research on spherically expanding flames has mainly been performed with a combustion tunnel [72, 81] and more recently with a combustion vessel. This latter device is often used to determine the burning velocity of different kinds of fuel at different pressures and initial temperatures and to determine the influence of the flame stretch on the burning velocity [15, 32, 47, 49, 54, 111]. The development of cellular structures on expanding flame fronts is one of the other interesting research areas in which combustion vessels are used [16, 44]. Most of the recent theoretical and numerical studies on these flames focus on predicting the burning velocity and the flame stretch rate, and on relations between these two parameters. In these studies, the chemistry is taken into account by using different reaction mechanisms, such as single-step reaction mechanisms [3, 73], reduced reaction mechanisms [17], and more detailed mechanisms [5, 63, 74]. In this thesis, the laminar spherical and cylindrical flames are studied numerically and analytically in order to understand how the burning and flame propagation velocities behave through the flames and how stretch affects these flames. In order to reduce the computational costs, a so-called flamelet model was developed to simulate these flames instead of solving the equations that govern combustion directly. In contrast to spherically expanding flames, the steady, cylindrical, and imploding flames are not often encountered in real situations and are therefore mainly studied to understand more about the behaviour of flames in general.

Expanding flames in a turbulent flow field are encountered in many industrial and consumer devices such as in spark-ignition engines [75, 105]. An example of such an engine is a petrol car engine. In a spark-ignition engine, the mixture of fuel and oxidizer is ignited by a spark discharge. After ignition, a laminar flame kernel grows rapidly into a turbulent expanding flame. Extensive experimental research has been performed on expanding flames in such engines [98]. Due to the high computational costs, numerical research is mostly restricted to simpler geometries and smaller domains than those of a real spark-ignition engine [2, 4, 34, 52]. However, these studies are a crucial first step in understanding combustion processes in real engines. With this knowledge, strategies can be found to increase the engine efficiency and to reduce the emission of pollutants. In this thesis, freely expanding flames in a turbulent flow field are studied numerically to understand how they are influenced by various turbulent flow fields. A direct numerical simulation program was developed for this purpose.

The outline of this thesis is as follows. In the first chapter, the concepts and definitions of various combustion parameters are provided, the general equations for modelling flames are presented, the geometries of the different flames that are investigated are shown in more detail, and the choice of the flame models is explained. Chapter 2 starts with the theory behind the new flamelet model to simulate perfectly spherical and cylindrical flames. Subsequently, this flamelet
model is used to simulate these flames and several important combustion variables are analysed such as the burning velocity, the flame propagation velocity, the flame thickness, and the flame stretch rate. We published the flamelet model and the analysis of the corresponding results in [45] and we presented this model and the results at the Ninth International Conference on Numerical Combustion (April 7–10, 2002, Sorrento, Italy), at the Twenty-Ninth International Symposium on Combustion (July 21–26, 2002, Sapporo, Japan), and at the First European Combustion Meeting (October 25–28, 2003, Orléans, France). In Chapter 3, the relation between the burning velocity and the flame stretch rate is analysed in more detail. This chapter focuses on theoretical expressions that describe this relation and on the limited usefulness of separating the flame stretch rate in a flow straining part and a curvature part. We published this analysis in [46] and presented it at the Third Joint Meeting of the US Sections of the Combustion Institute (March 16–19, 2003, Chicago, United States of America). The next chapter deals with the direct numerical simulation of expanding flames in a turbulent environment. This chapter starts with the description of the numerical program that was developed to perform the direct numerical simulations. This program is described in terms of the discretisation of the combustion equations, the boundary conditions, the chemical reduction methods, and the properties of the mesh. Validation of this program is performed by comparing simulations on stretchless one-dimensional flames and strongly stretched spherically expanding flames to results from a one-dimensional numerical reference program. Subsequently, simulations on turbulent flames are shown. We presented the results obtained in this chapter at the 19th International Colloquium on the Dynamics of Explosions and Reactive Systems (July 27 – August 1, 2003, Hakone, Japan). Also, a publication based on this chapter is in preparation. The final chapter summarizes the main conclusions and gives some recommendations for future research.

I am greatly indebted to many of my colleagues, family, and friends for their support during the last four years. I would like to give an extra word of thanks to some people in particular. First of all, I want to specially thank my supervisor professor Philip de Goey who supported me throughout this research project, and who taught me the essentials on combustion and its basic physical principles. Also, professor Norbert Peters is gratefully acknowledged for giving me the opportunity to perform part of the underlying investigation under his supervision at the Rheinisch-Westfälische Technische Hochschule Aachen in Germany. Furthermore, professor Mico Hirschberg and professor Dirk Roekaerts have carefully read a draft of this thesis and their suggestions and comments helped me a great deal to improve the text. My colleagues in both Eindhoven and Aachen made my stay at these universities a very pleasant one. Especially, I wish to thank Happy Bongers with whom I shared my office in Eindhoven and with whom I had many pleasant discussions about various subjects, and Rob Bastiaans and Jeroen van Oijen who offered their valuable help in the research as described in Chapter 4. Also, the continuous support of my father, grandmother, and two brothers meant a great deal to me. As usual, the final person to be mentioned here is the most important one. In my case, this is Werner, who was always there for me during
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Eindhoven, October 2003

Gemneke Groot
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1
General introduction

1.1 Introduction

The subject of this thesis is the modelling of spherical and cylindrical premixed methane-air flames. More specifically, these flames are steady, expanding, and imploding laminar flames, and expanding turbulent flames. The main goal is to investigate the behaviour of these flames by using two distinct numerical methods that were developed in this thesis. In this chapter, some more fundamental information about premixed flames is given, because this background is required for reading the remaining chapters.

Section 1.2 provides the most important concepts and definitions of premixed flames that are extensively used in the following chapters. Subsequently, an elaboration of the governing combustion equations is given in Section 1.3. These combustion equations are the basis of the numerical methods that were developed to simulate spherical and cylindrical flames. More detailed background information about this kind of flames and the applied numerical methods is given in Section 1.4.

1.2 Concepts and definitions

Premixed flames can be adequately described with the proper definition of the inner flame structure, the kinematics of the flame and flow, the stretching of a flame, and the flame thickness. These flame properties and their definitions are discussed in this section.

A premixed flame can be considered to consist of three layers, whose properties are discussed in Section 1.2.1. The kinematics of the flame is determined by the burning velocity, flame propagating velocity, flow velocity, and their mutual relations (cf. Section 1.2.2). Another fundamental concept is flame stretching, which is quantitatively described in Section 1.2.3. Finally, the thickness of a flame can be computed using different expressions which lead to different values. The best choice depends on the combustion model that is used and the particular situation. In Section 1.2.4, three commonly used expressions are discussed.
1.2.1 Inner flame structure

A schematic representation of the inner structure of a premixed flame is shown in Fig. 1.1. A large amount of fuel and oxidizer is present at the unburnt side of the flame, whereas this amount is significantly reduced at the burnt side. The opposite holds for carbon dioxide. This flame region can be divided into three zones. Going through the flame from the side of the unburnt gases to that of the burnt gases, one encounters the preheat zone first, then the inner layer, and finally the oxidation layer. The preheat zone can be considered as chemically inert. In this zone, the unburnt gases are heated up by heat transport from the reaction layer. The reaction layer can be divided into the inner and oxidation layer. In the inner layer, or fuel-consumption layer, the fuel is consumed and converted into hydrogen and carbon monoxide, and the radicals are depleted by chain-breaking reactions. The oxidation layer is located downstream of the reaction layer in which the oxidation takes place of hydrogen and carbon monoxide to water and carbon dioxide. The effect of the oxidation layer on the other two upstream layers is weak.

The thickness of the inner layer is small compared to the thickness of the preheat zone and the oxidation layer. The inner layer for methane-air flames under atmospheric pressure is typically around ten times smaller than the preheat zone and three times smaller than the oxidation layer [83]. Therefore, for practical use, the inner layer is commonly regarded as infinitely thin. In literature, the position of this infinitely thin inner layer is determined in several ways which lead to slightly different positions in the flame. The most commonly used positions in the flame are the ones where the heat release reaches its maximum, where the tangent of the temperature profile is the steepest, and where the concentration
of hydrogen H$_2$ or formyl radical HCO reaches its maximum. In this thesis, the maximum heat release is used. This choice is not crucial, because the different choices lead to positions in the flame that are close to each other.

### 1.2.2 Velocities

A premixed flame can be defined as a region in space where the combustion variables vary. These combustion variables are the temperature, density, and species mass fractions. A flame surface inside this flame can be defined as an iso-surface of a certain combustion variable that increases or decreases continuously going from the unburnt to the burnt side of the flame (cf. Fig. 1.1). The choice of the combustion variable is not crucial for the flame analysis in this thesis, because the iso-surfaces obtained from the different combustion variables have almost parallel orientations and they move with almost identical speed. In this thesis, the choice was made to define the flame surfaces as iso-surfaces of the density in Chapter 2 and of the temperature in Chapter 4.

The propagation of a flame surface in the normal direction and relative to the gases into the unburnt mixture is due to the chemical reactions that occur and is called the local laminar burning velocity $s_L$. The velocity at which the flame surfaces move is called the flame propagation velocity $v_f$. This flame propagation velocity $v_f$ can be obtained from the local balance between the flow velocity $v$ and the laminar burning velocity $s_L$:

$$v_f = v + s_L n_f,$$  \hfill (1.1)

in which $n_f$ is the unit vector normal to the flame surface pointing towards the unburnt gases. Fig. 1.2 shows these velocities at an iso-surface of the combustion variable $Y$.

The laminar burning velocity, flow velocity, and flame propagation velocity can vary between flame surfaces and along a flame surface. The structure of laminar flames hardly changes while moving, which indicates that the flame propagation velocity field is almost uniform. However, due to the expansion of the...
gases, the flow velocity field and the laminar burning velocity field vary significantly through the flame. It is often more convenient to use a variable that varies only slightly through the flame. Therefore, in this thesis, the mass burning rate is used instead of the laminar burning velocity. The mass burning rate $m$ is defined as the laminar burning velocity $s_L$ multiplied with the density $\rho$:

$$ m := \rho s_L. $$

In the case of a flat stretchless flame, this mass burning rate is uniform throughout the flame.

### 1.2.3 Flame stretch

Stretching of a flame can lead to variations of the flame behaviour compared to a flat flame with a purely one-dimensional flow and transport. This concept of flame stretch was first introduced by Karlovitz et al. [53]. The main causes of these stretch effects are the curvature of a moving flame surface and a gradient of the flow along this surface. A variable that takes the stretch effects into account is the flame stretch rate $K$. The definition of this rate and different theoretical expressions for it are discussed in this section.

The generally accepted definition of the flame stretch rate $K_A$ was first suggested by Williams [114]:

$$ K_A := \frac{1}{A} \frac{dA}{dt}. $$

This stretch rate represents the fractional area change of a small flame surface element $A$ which moves with the flame propagation velocity. From this definition in Eq. (1.3), more manageable expressions have been derived. In 1979, Buckmaster [19] derived an expression in cartesian coordinates. This expression, however, depends on the choice of the coordinate system and the representation of the flame surface. Therefore, Matalon [71] deduced an invariant formulation based on the results of Buckmaster. Subsequently, Chung and Law [23] derived a more concise expression for the flame stretch rate using the invariant formulation:

$$ K_A = \nabla \cdot v_t + \left( v_f \cdot n_f \right) \nabla \cdot n_f, $$

with $v_t$ the flow velocity in the tangential direction of the flame surface.

The first term on the right-hand side of Eq. (1.4) represents the contribution to flame stretch due to a nonuniform flow along the flame surface. An example of a flame which experiences only this straining effect $\nabla \cdot v_t$ is a stagnation flame. The second term represents the contribution to flame stretch due to the change in time of curvature of a flame surface. An example of such a flame which does not experience the straining effect $\nabla \cdot v_t$ but only the curvature effect $(v_f \cdot n_f) \nabla \cdot n_f$ is a perfectly spherical expanding flame. This means that a flame is not stretched when the flame front is not moving ($v_f = 0$), in combination with a gas flow which is perpendicular to the flame front, as in the case of a steady spherical
### 1.2 Concepts and Definitions

Flame or a flat flame with a uniform gas flow along the flame surface. The above mentioned is illustrated in Fig. 1.3 together with an example of a flame which contains both stretch effects.

The definition of the flame stretch rate in Eq. (1.3) is valid for infinitely thin flames, because the stretch field is defined on a flame sheet that characterizes the location of the flame. De Goey and ten Thije Boonkkamp [39] have extended expression (1.3) to a definition for the flame stretch rate that takes the flame thickness into account, by defining a stretch field inside the flame volume instead. They considered a small arbitrary control volume in the flame region \( V(t) \) to move with the local flame propagation velocity and have written the mass \( M(t) \) contained in this control volume as:

\[
M(t) = \int_{V(t)} \rho \, dV.
\]  

By applying Reynolds’ transport theorem [22], the rate of change of this mass in time is given by

\[
\frac{dM}{dt} = \int_{V(t)} \left( \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}_f) \right) \, dV.
\]  

Figure 1.3: Overview of the different stretch effects on flames. The continuous lines represent the flame front. The continuous and the dashed arrows represent the velocity of the gases and of the flame front, respectively.
According to de Goey and ten Thije Boonkkamp, this rate of change of mass \( \frac{dM}{dt} \) is related to the flame stretch rate \( K_M \) as:

\[
\frac{dM}{dt} = \int_{V(t)} \rho K_M \, dV.
\] (1.7)

The new definition of the flame stretch rate that takes the flame thickness into account is derived by dividing Eq. (1.7) by Eq. (1.5) and by considering an infinitesimal small control volume \( V(t) \rightarrow 0 \):

\[
K_M := \frac{1}{M} \frac{dM}{dt}.
\] (1.8)

The difference with the definition of the flame stretch rate in Eq. (1.3) is that the definition in Eq. (1.8) is based on the mass in the flame region instead of on the infinitely thin flame sheet.

A more manageable expression for this stretch rate can be derived by combining Eqs. (1.6) and (1.7) and the relation \( \frac{d}{dt} = \frac{\partial}{\partial t} + v_f \cdot \nabla \rho \):

\[
K_M = \nabla \cdot v_f + \frac{1}{\rho} \frac{d\rho}{dt}.
\] (1.9)

An invariant expression in terms of the local fluid velocity and the shape of the flame front is obtained by splitting the velocity of the flame surface into two components. One component normal to the local flame surface \( v_{nf}^f \) and the other one tangential to the surface \( v_{tf}^f \) which is equal to the tangential component of the fluid velocity \( v^f \):

\[
v_f = v_{nf}^f + v_{tf}^f = \left( v_f \cdot n_f \right) n_f + v^f.
\] (1.10)

Substitution of these components of the flame propagation velocity into Eq. (1.9) yields the invariant expression for the flame stretch rate:

\[
K_M = \nabla \cdot v^f + \left( v_f \cdot n_f \right) \nabla \cdot n_f + n_f \cdot \nabla \left( v_f \cdot n_f \right) + \frac{1}{\rho} \frac{d\rho}{dt}.
\] (1.11)

The first two terms on the right-hand side of Eq. (1.11) are the same as in the expression for the flame stretch rate in Eq. (1.4). The two extra terms are due to the fact that the thickness of the flame is taken into account. These terms vanish when an infinitely thin flame is considered. The third term of Eq. (1.11) arises from flame thickness variations due to different flame propagation velocities of different flame contours. The fourth term contributes to the stretch rate due to density variations along the flame iso-contours in case the density is not chosen as the progress variable. De Goey et al. [41] have shown that in case of a steady stoichiometric methane-air Bunsen flame the contributions to the stretch rate of these last two terms are quite small in comparison with the first two terms of Eq. (1.11).
The flame stretch rate is a very important parameter of the propagating spherical and cylindrical flames that are investigated in this thesis. Therefore, this rate and its influence on these flames are studied in more detail in the next chapters. In Chapter 2, the expressions (1.4) and (1.11) are used directly in the flamelet model and, in Chapter 3, the relation between the flame stretch rate and the mass burning rate is elaborated.

1.2.4 Flame thickness

An important length scale in premixed flames is the flame thickness. Several definitions of this flame thickness are in use and three of these definitions are discussed here.

The classical definition of the flame thickness $\delta_f$ is based on a geometrical approach. In this case, the flame thickness is the interval of the steepest tangent to the temperature profile between the unburnt and adiabatic temperature (cf. Fig. 1.4):

$$
\delta_f := \frac{T_{ad} - T_u}{(dT/ds)|_{T_{il}}},
$$

(1.12)

with $T$ the temperature, $s$ the coordinate perpendicular to the flame surface, and the subscripts $ad$, $il$, and $u$ denoting the adiabatic situation, the inner layer, and the unburnt gases, respectively.

Another definition that is commonly used is obtained by considering the flame thickness as a diffusive length scale. By assuming that the preheat zone is chemically inert, Göttgens et al. [43] have derived the following expression for the flame thickness:

$$
\delta_f := \frac{(\lambda/\varepsilon_p)|_{T_{il}}}{(\rho s_L)_u},
$$

(1.13)

where the product of the density and the laminar burning velocity $\rho s_L$ is evaluated in the unburnt gases $u$ and where the ratio of the heat conductivity over the heat capacity at constant pressure $\lambda/\varepsilon_p$ is evaluated at the inner layer temperature $T_{il}$. From a geometrical point of view, the definition in Eq. (1.13) is a measure for the width of the preheat zone of the flame.

In the definitions (1.12) and (1.13), the temperature of the inner layer is defined as the temperature at which the tangent of the temperature profile is the steepest [85]. The relationship between the definitions (1.12) and (1.13) is approximately:

$$
\delta_f^{(1.12)} \approx \delta_f^{(1.13)} \left( \frac{T_{ad} - T_u}{T_{il} - T_u} \right),
$$

(1.14)

De Goey and ten Thije Boonkkamp [40] have derived an expression for the flame thickness from a flamelet description:

$$
\delta_f := \int_{s_u}^{s_b} \left( \frac{\rho - \rho_u}{\rho_b - \rho_u} \right) ds,
$$

(1.15)
with the subscripts $u$ and $b$ denoting the unburnt and burnt side of the flame, respectively. In this definition not only the preheat zone is taken into account, but also the inner layer and part of the oxidation layer depending on the choice of the burnt flame boundary $s_b$.

In order to get an idea of the size of a flame, the flame thickness of a stoichiometric methane-air flame at a pressure of 1.0 atm and a temperature of the unburnt gases of 300 K is computed for each definition. The flame thickness based on the geometrical approach in Eq. (1.12) becomes 0.449 mm. Eq. (1.13) results in a flame thickness of 0.175 mm and Eq. (1.15) in 0.253 mm, whereby the burnt flame boundary is located at the position in the flame where the heat release is three times smaller than its maximum.

The choice which definition of the flame thickness to use is depending on the purpose of the model. The geometrical approach in Eq. (1.12) and the definition of de Goey and ten Thije Boonkkamp in Eq. (1.15) can be obtained from the temperature and density profiles of experimental or numerical data, but these definitions are difficult to use for theoretical purposes. For these theoretical purposes, Eq. (1.13) is easier to use as for example in Chapter 3. In Chapter 2, the geometrical definition in Eq. (1.12) and the definition of de Goey and ten Thije Boonkkamp in Eq. (1.15) are used to compute the thickness of steady, expanding, and imploding flames.

### 1.3 Governing equations

Combustion processes can be described by the conservation equations from the fluid dynamics and from the chemical composition. In fluid dynamics, the non-reacting flows are described with the conservation equations of mass, momentum, and energy. In three-dimensional compressible flows, these five equations contain five primitive variables, which are the density, the velocities in three directions, and the energy or temperature. To describe combustion processes, the mass conservation equations for the various chemical species have to be solved.
1.3 Governing equations

Additionally to obtain the mass fractions of the species involved.

The general conservation equations of mass, momentum, energy, and chemical components are presented in Section 1.3.1. In these conservation equations, the enthalpy and pressure are dependent on the primitive variables by means of two state equations, which are presented in Section 1.3.2. The conservation equations require modelling of the viscous stress tensor, the diffusion velocities, the heat flux, and the chemical source terms. These models are discussed in Section 1.3.3. By substitution of these models into the conservation equations, a set of combustion equations is obtained that is used in the remainder of this thesis (cf. Section 1.3.4).

1.3.1 General conservation equations

Conservation of mass is given by the continuity equation:

\[ \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = 0, \quad (1.16) \]

and conservation of momentum is described by the Navier-Stokes equations:

\[ \frac{\partial (\rho \mathbf{v})}{\partial t} + \nabla \cdot (\rho \mathbf{v} \mathbf{v}) = \mathbf{g} - \nabla \cdot \mathbf{P}, \quad (1.17) \]

with \( \mathbf{g} \) the gravitational acceleration field and \( \mathbf{P} \) the stress tensor, which is defined as \( \mathbf{P} = p \mathbf{I} - \mathbf{\tau} \), with \( p \) the hydrostatic pressure, \( \mathbf{I} \) the unit tensor, and \( \mathbf{\tau} \) the viscous stress tensor.

The transport equation for the conservation of energy is

\[ \frac{\partial (\rho e)}{\partial t} + \nabla \cdot (\rho e \mathbf{v}) = \mathbf{v} \cdot \mathbf{g} - \nabla \cdot \mathbf{q} - \nabla \cdot (\mathbf{P} \mathbf{v}) , \quad (1.18) \]

where \( \mathbf{q} \) is the heat flux field and \( e \) the energy density, which can be written as \( e = \hat{u} + \frac{1}{2} |\mathbf{v}|^2 \), \( \hat{u} \) being the internal energy density and \( \frac{1}{2} |\mathbf{v}|^2 \) the kinetic energy density. For modelling of combustion processes, the energy equation in Eq. (1.18) is commonly written in a different form using the enthalpy density \( h \), which is related to the internal energy density \( \hat{u} \) by \( h = \hat{u} + p/\rho \):

\[ \frac{\partial (\rho h)}{\partial t} + \nabla \cdot (\rho h \mathbf{v}) = \mathbf{v} \cdot \mathbf{g} - \nabla \cdot \mathbf{q} + \mathbf{\tau} : (\nabla \mathbf{v}) + \frac{dp}{dt} , \quad (1.19) \]

in which \( dp/dt := \partial p/\partial t + \mathbf{v} \cdot \nabla p \).

For every species in a chemically reacting flow, a mass-balance equation can be introduced:

\[ \frac{\partial \rho_i}{\partial t} + \nabla \cdot (\rho_i \mathbf{v}_i) = \dot{\rho}_i , \quad (1.20) \]

where \( \rho_i \) and \( \mathbf{v}_i \) are the mass density and particular velocity of species \( i \), respectively. In contrast to the total mass conservation equation (1.16), this equation
has a source term $\dot{\rho}_i$ which describes the rate of change of the mass of species $i$ due to chemical reactions. The velocity of species $i$ can be written as $v_i = v + V_i$, with $V_i$ the diffusion velocity which is the deviation of the velocity of each species from the average flow velocity $v$. By substituting this relation for the velocity of species $i$ and by using the definition of the mass fraction of each species $Y_i = \rho_i/\rho$, Eq. (1.20) can be written in a form that is more commonly used:

$$\frac{\partial (\rho Y_i)}{\partial t} + \nabla \cdot (\rho v Y_i) = -\nabla \cdot (\rho V_i Y_i) + \dot{\rho}_i. \quad (1.21)$$

A detailed derivation of the conservation equations described in this section can be found in [11, 58, 115].

### 1.3.2 State equations

In the conservation equations presented in Section 1.3.1, the pressure and the enthalpy can be obtained from the density, the temperature, and the species mass fractions by the thermal equation of state and the caloric equation of state, respectively. These equations are presented here.

The thermal equation of state is the so-called ideal gas law, which relates the pressure to the density, the temperature, and the species mass fractions. All species in the flow are considered to behave as an ideal gas. Therefore, the ideal gas law for the pressure for each species $p_i$ reads:

$$p_i = n_i RT = n X_i RT, \quad (1.22)$$

in which $R$ is the universal gas constant and $X_i = n_i/n$ the species mole fraction, with $n_i$ and $n$ the molar concentration of the species $i$ and the total gas mixture, respectively. The species mole fraction $X_i$ is related to the species mass fraction $Y_i$ by

$$X_i = Y_i \frac{\bar{M}}{M_i}, \quad (1.23)$$

where $M_i$ is the molar mass of species $i$ and $\bar{M}$ the mean molar mass. Using Eq. (1.23) and knowing that $\rho = n\bar{M}$, Eq. (1.22) can be rewritten as

$$p_i = \rho RT \frac{Y_i}{M_i}. \quad (1.24)$$

The hydrostatic pressure $p$ is equal to the sum of the partial pressures $p_i$ of all species:

$$p = \sum_{i=1}^{N_s} p_i = \rho RT \sum_{i=1}^{N_s} \frac{Y_i}{M_i}, \quad (1.25)$$

with $N_s$ the number of species.
1.3 Governing equations

The caloric equation of state relates the enthalpy density \( h \) to the temperature \( T \) and the species mass fractions \( Y_i \) as:

\[
h = \sum_{i=1}^{N_s} Y_i h_i, \quad \text{with} \quad h_i = h_i^* + \int_{T^*}^{T} c_{p_i}(T) \, dT,
\]

where \( h_i \) is the enthalpy density of species \( i \), \( h_i^* \) the species enthalpy density of formation at the reference temperature \( T^* \), and \( c_{p_i} \) the specific heat of species \( i \) at constant pressure. This specific heat \( c_{p_i} \) can be tabulated in polynomial form, cf. [55]:

\[
\frac{c_{p_i}}{R} = \sum_{n=1}^{5} b_{n,i} T^{n-1}.
\]

The specific heat at constant pressure of the total gas mixture \( c_p \) is dependent on both the species heat capacities \( c_{p_i} \) and the species mass fractions \( Y_i \):

\[
c_p = \sum_{i=1}^{N_s} Y_i c_{p_i}.
\]

1.3.3 Transport and chemistry models

First, transport models are presented for the viscous stress tensor \( \tau \), the diffusion velocities \( V_i \), and the heat flux \( q \). Subsequently, a chemistry model is elaborated for the chemical source terms \( \rho_i \).

Transport models

The gas mixture is assumed to behave as a Newtonian fluid. Therefore, the viscous stress tensor \( \tau \) can be modelled with Stokes’ law of friction, cf. [99]:

\[
\tau = \mu \left( \nabla v + (\nabla v)^T - \frac{2}{3} (\nabla \cdot v) \mathcal{I} \right),
\]

with \( \mu \) the dynamic viscosity of the mixture, which is related to the kinematic viscosity \( \nu \) as \( \mu = \rho \nu \). In the case of aerodynamic flows, the following formula based on Sutherland’s theory of viscosity can be used to compute this dynamic viscosity, cf. [99]:

\[
\mu = C_1 \frac{T^2}{T + C_2},
\]

with \( C_1 = 1.458 \cdot 10^{-6} \, \text{kg m}^{-1} \text{s}^{-1} \text{K}^{-\frac{1}{2}} \) and \( C_2 = 110.4 \, \text{K} \). Eq. (1.30) can also be used for the flame computations in this thesis, because the flames considered here mainly consist of air.
The diffusion velocity field $V_i$ can be obtained by solving the following system of equations, cf. [115]:

$$\nabla X_i = \sum_{p=1}^{N_s} \frac{X_i X_p}{D_{ip}} (V_p - V_i) \quad \text{for} \quad i = 1, N_s,$$

(1.31)

in which the influence of the pressure gradients and the diffusion of mass due to temperature gradients are neglected, and where $D_{ip}$ is the binary mass diffusion coefficient of species $i$ into species $p$. These equations are usually referred to as the Stefan-Maxwell equations. The computational costs to solve the system (1.31) are very high. Therefore, a simplified approach based on Fick’s law is commonly used to obtain the diffusion velocities, cf. [68]:

$$V_i = -\frac{D_{im}}{Y_i} \nabla Y_i,$$

(1.32)

with $D_{im}$ the diffusion coefficient of species $i$ into the mixture $m$. This equation is proven to perform well in simulations of methane-air flames, because nitrogen is abundant in that case.

For the heat flux $q$, an expression is used that contains the heat transport due to conduction and mass diffusion, cf. [115]:

$$q = -\lambda \nabla T + \rho \sum_{i=1}^{N_s} V_i Y_i h_i,$$

(1.33)

with $\lambda$ the thermal conductivity of the gas mixture. The heat flux $q$ in Eq. (1.33) can be rewritten in terms of the enthalpy and the species mass fraction instead of the temperature and diffusion velocities by substitution of Eqs. (1.26), (1.28), and (1.32) into Eq. (1.33):

$$q = -\frac{\lambda}{c_p} \nabla h - \frac{\lambda}{c_p} \sum_{i=1}^{N_s} \left( \frac{1}{Le_i} - 1 \right) h_i \nabla Y_i,$$

(1.34)

in which $Le_i$ represents the Lewis number of species $i$, which is the ratio of the thermal conduction to the species diffusion:

$$Le_i = \frac{\lambda}{c_p \rho D_{im}}.$$

(1.35)

In order to save computational costs, it is assumed in this thesis that the Lewis numbers are spatially uniform. The use of uniform Lewis numbers is justified, because the Lewis numbers change only slightly through the methane-air flames. Furthermore, these uniform Lewis numbers have been proven to be well suited for modelling methane-air flames (cf. [90]). The ratio of the thermal conductivity over the heat capacity at constant pressure $\lambda/c_p$ in Eq. (1.34) is dependent on
the temperature and the composition of the gas mixture. Here, a simplified relation is used that is dependent on the temperature only, in order to reduce the computational costs, cf. [102]:

\[
\frac{\lambda}{\varepsilon_p} = 2.58 \times 10^{-5} \left( \frac{T}{298} \right)^{0.7}.
\]  

The model in Eq. (1.36) is only valid for flames that contain large quantities of nitrogen, because this model is obtained from fitting to data of methane-air flames.

**Chemical model**

The global reaction for the complete combustion of methane and air is:

\[
\text{CH}_4 + 2\text{O}_2 \rightarrow \text{CO}_2 + 2\text{H}_2\text{O}.
\]  

This reaction represents the overall reaction of the many elementary reactions which occur on a molecular level in the combustion process. The general form of these elementary reactions is:

\[
\sum_{i=1}^{N_r} y_i' A_i = \sum_{i=1}^{N_r} y_i'' A_i \quad \text{for} \quad l = 1, N_r,
\]  

where \(y_i'\) and \(y_i''\) are the molar stoichiometric coefficients of species \(i\) in reaction \(l\), \(N_r\) the number of reactions, and \(A_i\) the chemical symbol for species \(i\) as for example CO, O, CH, and NH.

In order to describe combustion processes accurately, the chemical kinetics of the elementary reactions have to be known. These chemical kinetics are incorporated into the combustion computations through the chemical source terms \(\dot{\rho}_i\) in the mass-balance equations (1.21):

\[
\dot{\rho}_i = M_i \sum_{l=1}^{N_r} (y_i' - y_i'') Q_l,
\]  

in which \(Q_l\) is the reaction rate for an elementary reaction \(l\):

\[
Q_l = k_f^l \prod_{i=1}^{N_r} [A_i]^{y'_i} - k_b^l \prod_{i=1}^{N_r} [A_i]^{y''_i},
\]  

where \([A_i] = \rho Y_i / M_i\) represents the molar concentration of species \(A_i\), \(k\) is the reaction rate coefficient, the subscript \(l\) denotes the reaction index, and the superscripts \(f\) and \(b\) refer to the forward and backward reactions, respectively. The reaction rate coefficient of a forward elementary reaction in Eq. (1.40) is usually written in an Arrhenius form:

\[
k_f^l = A_f^l \theta_f^l e^{-\frac{E_f^l}{RT}},
\]  

in which \(A_f^l\) is the pre-exponential factor for the \(l\)-th forward reaction.
with $A_c$ the pre-exponential constant, $\beta$ the temperature exponent, and $E_a$ the activation energy. These constants are mostly obtained experimentally. The backward reaction rates can be computed from the forward rates through the equilibrium constants $K_{ci} = k_i^f/k_i^b$, cf. [56].

Many authors have selected elementary reactions and measured the corresponding rate data. Tables that contain this information are called elementary reaction mechanisms. A mechanism that is often used for simulations of methane-air flames is GRI-mech 3.0 which is compiled by the Gas Research Institute [14]. It comprises 325 reactions among 53 chemical species. An example of a smaller reaction mechanism can be found in [102]. It contains 25 reactions and 16 species. This mechanism is called a skeletal mechanism, because it is considered to contain a minimal subset of the complete mechanism for methane-air flames. This skeletal mechanism and GRI-mech 3.0 are used in Chapters 2 and 4, respectively.

### 1.3.4 Combustion equations

The total set of combustion equations that is used in this thesis consists of the continuity equation as it is shown in Eq. (1.16), the Navier-Stokes equations, the enthalpy or temperature equation, and the mass-balance equations. These equations are obtained by inserting the models described in Section 1.3.3 into the conservation equations in Section 1.3.1. The resulting equations are shown below. Together with the two state equations in Section 1.3.2, the density, the velocities, the temperature, and the species mass fractions can be computed. Notice that the mass balance equations have to be solved for $N_s - 1$ species instead of $N_s$ species, because the mass fraction and the corresponding Lewis number of one of the species can be computed with the following constraints:

$$
\sum_{i=1}^{N_s} Y_i = 1 \quad \text{and} \quad \sum_{i=1}^{N_s} V_i Y_i = 0.
$$

(1.42)

The abundant species is commonly chosen for this species, which is nitrogen here.

After substitution of the model for the viscous stress tensor that is presented in Eq. (1.29), the Navier-Stokes equations (1.17) become:

$$
\frac{\partial (\rho \mathbf{v})}{\partial t} + \nabla \cdot (\rho \mathbf{v} \mathbf{v}) = \rho \mathbf{g} - \nabla p \\
+ \nabla \cdot \left( \mu \left( \nabla \mathbf{v} + (\nabla \mathbf{v})^T - \frac{2}{3} (\nabla \cdot \mathbf{v}) \mathbf{I} \right) \right).
$$

(1.43)

By inserting Eq. (1.34) into Eq. (1.19) and by using Eq. (1.35), the enthalpy equation (1.19) becomes:

$$
\frac{\partial (\rho h)}{\partial t} + \nabla \cdot (\rho \mathbf{v} h) = \nabla \cdot \left( \frac{\lambda}{\varepsilon_p} \nabla h \right) + \nabla \cdot \left( \frac{\lambda}{\varepsilon_p} \sum_{i=1}^{N_s} \left( \frac{1}{Le_i} - 1 \right) h_i \nabla Y_i \right) \\
+ \rho \mathbf{v} \cdot \mathbf{g} + \mathbf{\tau} : (\nabla \mathbf{v}) + \frac{dp}{dt}.
$$

(1.44)
In the derivation of the flamelet model in Chapter 2, the very small contributions of the enthalpy production are neglected. These contributions are the volumetric heat sources $\rho v \cdot g$, the viscous effects $\mathbf{\tau} : (\nabla v)$, and the pressure variations $\frac{dp}{dt}$.

In the DNS program described in Chapter 4, the conservation of energy is used in terms of the temperature instead of in terms of the enthalpy. This temperature equation can be obtained from the enthalpy equation (1.19) using Eqs. (1.21), (1.26), (1.32), (1.34), and (1.35):

$$c_p \frac{\partial (\rho T)}{\partial t} + c_p \nabla \cdot (\rho v T) = \nabla \cdot (\lambda \nabla T) - \sum_{i=1}^{N_s} h_i \rho_i + \sum_{i=1}^{N_s} \rho D_{i,\text{int}} c_{p,i} \nabla T \cdot \nabla Y_i$$

$$+ \rho v \cdot g + \mathbf{\tau} : (\nabla v) + \frac{dp}{dt},$$

(1.45)
in which the chemistry model explained in Section 1.3.3 is used for the chemical source term $\dot{\rho}_i$. Although the last four terms on the right-hand side of Eq. (1.45) are small compared to the other terms, these four terms are not neglected in Chapter 4.

The mass-balance equation for each species $i$ becomes:

$$\frac{\partial (\rho Y_i)}{\partial t} + \nabla \cdot (\rho v Y_i) = \nabla \cdot \left( \frac{\Lambda}{Le} c_p \nabla Y_i \right)$$

$$+ M_i \sum_{l=1}^{N_s} A_{i,l} r_{i,l}^{\alpha} e^{-\frac{E_i}{RT}} (v_i'' - v_i') \left( \prod_{l=1}^{N_s} \left( \frac{\rho Y_i}{M_i} \right)^{v_i''} - \frac{1}{K_i} \prod_{l=1}^{N_s} \left( \frac{\rho Y_i}{M_i} \right)^{v_i'} \right)$$

(1.46)
after inserting the model for the diffusion velocities (cf. Eq. (1.32)) and the chemistry model (cf. Eqs. (1.39), (1.40), and (1.41)) into Eq. (1.21) and using the definition of the Lewis number in Eq. (1.35).

1.4 Spherical and cylindrical flames

In this thesis, perfectly spherical and cylindrical flames and expanding turbulent flames are considered. In Section 1.4.1, the geometries of these laminar and turbulent flames are shown. These flames are simulated using two distinct methods. The background to the choice of these methods is explained in Section 1.4.2.

1.4.1 Schematic representations

The main focus of this investigation lies on perfectly spherical and cylindrical flames without instabilities on the flame surface. These flames can expand, implode, or be motionless. Schematic representations of the expanding and imploding flames are shown in Fig. 1.5. Besides these laminar flames, also expanding flames in a turbulent environment are simulated. A schematic representation of such an expanding flame is shown in Fig. 1.6.
Fig. 1.5: Schematic representations of an expanding (a) and an imploding (b) flame. The continuous lines represent the reaction layer, the dashed lines the front of the preheat zone, and $\hat{n}_f$ the component of the normal unit vector in the radial direction.

Fig. 1.5 shows the perfectly spherical and cylindrical flames as simulated and discussed in Chapters 2 and 3. These flames are rotationally symmetric and can therefore be described with the radial coordinate $r$ only. The most important variables for this study are the gas velocity in the radial direction $v^r$, the laminar burning velocity $s_L$, the flame propagation velocity in the radial direction $v_f^r$, the flame thickness $\delta_f$, and the flame stretch rate. Besides unsteady flames, also steady flames are investigated. This implies that an imaginary sink for burnt gases or a source of unburnt gases is present at the centre of the expanding and imploding flames, respectively, to prevent the flame from moving.

Fig. 1.6 shows an expanding flame in a turbulent environment as simulated and discussed in Chapter 4. The gases are in turbulent motion and are considered to be both homogeneous and isotropic. The flame front is able to wrinkle and develop instabilities due to the turbulent motion of the flow and the thermodynamic expansion of the gases. The flames are described in three-dimensional space, because turbulence is essentially three-dimensional, and can not be described in two dimensions. A cartesian coordinate system $x = (x, y, z)$ is used. In Fig. 1.6, $v = (u, v, w)$ represents the gas velocity field and $v_f = (v_f^x, v_f^y, v_f^z)$ the flame propagation velocity field.

### 1.4.2 Methods

In this thesis, two distinct methods are used to simulate the flames: the flamelet method (cf. Chapter 2) and the Direct Numerical Simulation (DNS) method (cf.
Chapter 4). A new flamelet model is developed to study the inner structure of the perfectly spherical and cylindrical flames in detail, and a new DNS program is developed to study the behaviour of freely expanding flames in a turbulent flow field.

The main idea behind flamelet modelling is the decoupling of the fast and slow processes in the flame. This decoupling reduces the computational costs significantly in comparison with direct numerical simulations, without losing significant accuracy and, therefore, keeping it possible to examine the flames in detail. Flamelet modelling is increasingly used in turbulent combustion modelling (e.g. [86]). It is, however, not often used in laminar flame computations, although it can have major advantages (e.g. [76]). In the case of perfectly spherical and cylindrical flames, it is possible to evaluate the local flame stretch rate directly from its definition (cf. Section 1.2.3).

DNS solves the instantaneous combustion equations (cf. Section 1.3) directly without the requirement of simplifying models (cf. e.g. [7, 37, 52, 89, 97]). Due to the wide range of length and time scales in combustion and turbulence processes, DNS is extremely time consuming, especially when computing large domains and complex chemistry. Nevertheless, other less expensive methods such as Reynolds Averaged Navier Stokes (RANS) (cf. e.g. [18, 21, 59, 86, 104]) and Large Eddy Simulation (LES) (cf. e.g. [38, 90, 106, 113]) are not used here, because these methods require closure models which make the solutions less accurate. Furthermore, the goal here is to gather detailed information about the inner structure of the flame as a function of time, which is impossible to obtain using these methods, because the variables inside the flame are averaged. In order to reduce the com-

Figure 1.6: Schematic representation of an expanding flame in a turbulent environment. The continuous line represents the reaction layer, the dashed line the front of the preheat zone, and $\mathbf{n}_f$ the unit vector perpendicular to the flame surface.
putational costs of the very expensive DNS, only a small domain is considered here in combination with a single-step reaction model (cf. [60]) and a chemical reduction method called Flamelet-Generated Manifolds (cf. [79]) for the description of the chemistry.
Flamelet modelling of spherical and cylindrical flames

2.1 Introduction

The computational costs of directly solving the combustion equations presented in Section 1.3 are huge. The mass-balance equations (1.46) increase the number of nonlinear equations significantly, because even the combustion of simple hydrocarbon fuels involves a large number of species. These mass-balance equations are strongly coupled by the chemical source terms and the chemical reactions occur on time scales that vary several orders of magnitude, which implies that the set of equations is stiff. The solution of this set of nonlinear and stiff equations puts very high demands on the numerical algorithm and is, therefore, a very difficult and time-consuming task. In order to reduce these costs, different combustion models have been developed in the last decades. Very promising ones are the so-called flamelet models. Examples of different flamelet models are found in [33, 57, 80, 86, 101]. The main idea behind these models is the decoupling of the fast chemical kinetic processes in the flame from the slower flow and mixing processes. This is a valid approach in flames where the time scales of the inner flame structure are much smaller than the time scales of the movement of the flame itself, so that the inner flame structure can adapt itself instantaneously to the flame movement. This is the case in perfectly spherical and cylindrical flames as long as the flames are far from ignition or extinction.

In Section 2.2, a new flamelet model is presented, which was especially developed to simulate perfectly spherical and cylindrical flames (cf. Section 1.4). This flamelet model is based on the flamelet concept deduced by de Goey and ten Thije Boonkkamp [40]. However, they could not perform computations using this flamelet concept, because information about the flow and the progress variable in the complete flamelet structure is required to determine the local burning velocity of a flamelet. To overcome this difficulty, a new computational strategy was developed. This computational strategy and the numerical implementation of the flamelet model are explained in Section 2.3.
Subsequently, the numerical results obtained from the flamelet model are presented and elaborated. The variables of interest are the mass burning rate, the burning velocity, the flame propagation velocity, the gas velocity, the flame stretch rate, and the thickness of the flame. These combustion variables are analysed here to obtain a better understanding of their behaviour inside the spherical and cylindrical flames. Details on the numerical computations are described in Section 2.4. In the next section, the mass burning rate, burning velocity, flame propagation velocity, and gas velocity at various positions in the flame are studied and compared with each other and with experimental and numerical data. The flame stretch rates inside the flames are shown in Section 2.6 as well as the change of the flame stretch rate due to the movement of the flames. The change of the flame thickness due to expansion or implosion is shown and explained in the last section.

2.2 Flamelet model

The flamelet model consists of three parts, which are presented separately in this section. The first part describes the flame propagation (cf. Section 2.2.1), the second part the flow of the gases (cf. Section 2.2.2), and the last part the internal structure of the flame (cf. Section 2.2.3). The chemical kinetics are taken into account in the last part.

2.2.1 Flame propagation

The first part of the flamelet model describes the position of the flame front and its evolution in time using a field equation which governs a scalar field $\mathcal{Y}(x, t)$. $\mathcal{Y}$ can be the density $\rho$ or one of the species mass fractions $Y_i$ that obeys $|\nabla Y_i| \neq 0$. A flame surface in the flame front is defined as an iso-plane of $\mathcal{Y}$, which means that on this flame surface $\mathcal{Y}(x, t) = \text{constant}$. $\mathcal{Y}$ is constant when moving along an iso-plane of the flame surface, which means that the following kinematic equation holds:

$$\frac{d\mathcal{Y}}{dt} := \frac{\partial \mathcal{Y}}{\partial t} + \mathbf{v}_f \cdot \nabla \mathcal{Y} = 0.$$  (2.1)

The flame propagation velocity $v_f$ in Eq. (2.1) can be obtained from the local balance between the flow velocity $\mathbf{v}$ and the burning velocity $s_L$, cf. Eq. (1.1):

$$v_f = v + s_L n_f.$$  (2.2)

The unit vector normal to the flame surface $n_f$ in Eq. (2.2) can be expressed as:

$$n_f = \text{sign} \left( \mathcal{Y}_u - \mathcal{Y}_b \right) \frac{\nabla \mathcal{Y}}{|\nabla \mathcal{Y}|},$$  (2.3)

where the subscripts $u$ and $b$ refer to the unburnt and burnt side of the flame, respectively. Notice that other flamelet models usually employ the $G$-equation
[69,115] instead of Eq. (2.1). These two equations are related to each other. However, in the $G$-equation approach only the iso-level $G = G_0$ is followed, which represents the position of a single iso-plane in the flame front.

In this flamelet model, the progress variable is the density, which indicates that a flame surface in the flame front is defined as an iso-plane of the density. Furthermore, the density and the flame propagation velocity of the perfectly spherical and cylindrical flames vary in the radial direction only (cf. Section 1.4.1). Therefore, for these spherical and cylindrical flames, the kinematic equation in Eq. (2.1) reduces to:

$$\frac{\partial \rho}{\partial t} + v^r \frac{\partial \rho}{\partial r} = 0,$$

(2.4)

with $v_f^r$ the flame propagation velocity in the radial direction.

2.2.2 Flow of the gases

The second part describes the flow of the gases. For perfectly spherical and cylindrical flames, the gas velocity has a non-zero component in the radial direction only. Furthermore, this gas velocity depends on the local density field and the flame propagation. These dependencies can be described by the continuity equation (1.16) and the kinematic equation for the density (2.4). By combining these two equations, the following equation for the gas velocity in the radial direction $v_f^r$ is obtained:

$$\frac{1}{\sigma} \frac{\partial}{\partial r} (\sigma v^r) = \frac{s_L n_f^r}{\rho} \frac{\partial \rho}{\partial r},$$

(2.5)

in which $\sigma$ is a measure for the area on the flame surfaces through which transport takes place. For the perfectly spherical and cylindrical flames, $\sigma = r^2$ and $\sigma = r$, respectively. In the derivation of Eq. (2.5), the following velocity balance is used (cf. Eq. (1.1)):

$$v_f^r = v^r + s_L n_f^r,$$

(2.6)

where $n_f^r$ is the component of the normal unit vector in the radial direction (cf. Eq. (2.3)):

$$n_f^r = \text{sign} (\rho_u - \rho_b) \frac{\partial \rho/\partial r}{|\partial \rho/\partial r|},$$

(2.7)

which is $-1$ for flames with unburnt gases at the centre of the sphere or cylinder and $1$ for flames with burnt gases at the centre.

2.2.3 Internal structure of the flame

The last part of the flamelet model describes the internal structure of the flame, thereby taking the chemical kinetics into account. In this part, the conservation equations of mass (1.16), enthalpy (1.44), and mass fraction for each species (1.46) are rewritten in a flame-adapted coordinate system. The axes of this orthogonal system $\eta = (\xi, \zeta, \eta)$ are locally adapted to the contours of the progress variable $\rho$. 

A schematic illustration of a part of a three-dimensional flame is shown in Fig. 2.1, in which \( \rho_u \) and \( \rho_b \) represent two iso-planes at the unburnt and the burnt side of the flame, respectively. The scale factors in Fig. 2.1 define the arc lengths in the \( \eta \)-coordinate system: \( h_\xi = |\partial x/\partial \xi| \), \( h_\zeta = |\partial x/\partial \zeta| \), and \( h_\eta = |\partial x/\partial \eta| \). The corresponding unit vectors are given by \( \mathbf{e}_\xi = (1/h_\xi) \partial \mathbf{x}/\partial \xi \), \( \mathbf{e}_\zeta = (1/h_\zeta) \partial \mathbf{x}/\partial \zeta \), and \( \mathbf{e}_\eta = (1/h_\eta) \partial \mathbf{x}/\partial \eta \). In terms of the unit vector normal to the flame surface \( \mathbf{e}_\eta \), the velocities through the flame relate to each other in the following way:

\[
\mathbf{v}_f = \mathbf{v} - s_L \mathbf{e}_\eta. \tag{2.8}
\]

Notice that \( \mathbf{e}_\eta = -\mathbf{n}_f \) (cf. Eqs. (2.2) and (2.8)).

Using Eq. (2.8), the continuity equation (1.16), the enthalpy equation (1.44), and the mass-balance equation for each species (1.46) can be reformulated as:

\[
\nabla \cdot (\rho s_L \mathbf{e}_\eta) = -\left( \frac{\partial \rho}{\partial t} + \nabla \cdot \left( \rho \mathbf{v}_f \right) \right), \tag{2.9}
\]

\[
\nabla \cdot (\rho s_L h \mathbf{e}_\eta) - \nabla \cdot \left( \frac{\Lambda}{c_p} \nabla h \right) - \nabla \cdot \left( \frac{\Lambda}{c_p} \sum_{i=1}^{N} \left( \frac{1}{Le_i} - 1 \right) h_i \nabla Y_i \right)
= -h \left( \frac{\partial \rho}{\partial t} + \nabla \cdot \left( \rho \mathbf{v}_f \right) \right) - \rho \frac{dh}{dt}, \tag{2.10}
\]

and

\[
\nabla \cdot (\rho s_L \mathbf{Y}_i \mathbf{e}_\eta) - \nabla \cdot \left( \frac{\Lambda}{Le_i c_p} \nabla Y_i \right) - \rho_i = -Y_i \left( \frac{\partial \rho}{\partial t} + \nabla \cdot \left( \rho \mathbf{v}_f \right) \right) - \rho \frac{dY_i}{dt}, \tag{2.11}
\]

respectively. In Eqs. (2.9), (2.10), and (2.11), the first term on the right-hand side between parentheses represents the density multiplied with the flame stretch field \( K \) (cf. Eq. (1.9)). The terms \( dh/\partial t := \partial h/\partial t + \mathbf{v}_f \cdot \nabla h \) and \( dY_i/\partial t := \partial Y_i/\partial t + \mathbf{v}_f \cdot \nabla Y_i \) in Eqs. (2.10) and (2.11) describe the relative movement of the iso-surfaces.
of the enthalpy and the mass fractions with respect to the iso-surfaces of the progress variable $\rho$. For the spherical and cylindrical flames, these terms are small compared to the stretch terms (cf. [40]) and can therefore be neglected.

By defining the mass burning rate as $m := \rho s L$ and the area field as $\sigma := h_1 h_2$, Eqs. (2.9), (2.10), and (2.11) can be expressed in the flame-adapted coordinate system $\eta = (\xi, \zeta, \eta)$:

$$\frac{\partial}{\partial s} (\sigma m) = -\sigma p K,$$  \hspace{2cm} (2.12)

$$\frac{\partial}{\partial s} (\sigma m h) - \frac{\partial}{\partial s} \left( \sigma \frac{\lambda}{c_p} \frac{\partial h}{\partial s} \right) - \frac{\partial}{\partial s} \left( \sigma \frac{\lambda}{c_p} \sum_{i=1}^{N_r} \left( \frac{1}{L e_i} - 1 \right) h_i \frac{\partial Y_i}{\partial s} \right) = -\sigma p K h,$$  \hspace{2cm} (2.13)

and

$$\frac{\partial}{\partial s} (\sigma m Y_i) - \frac{\partial}{\partial s} \left( \sigma \frac{\lambda}{L e_i c_p} \frac{\partial Y_i}{\partial s} \right) - \sigma p_i = -\sigma p K Y_i,$$  \hspace{2cm} (2.14)

respectively, where $s$ represents the arc length perpendicular to the flame surface ($ds = h_0 d\eta$). Eqs. (2.12), (2.13), and (2.14) constitute the flamelet equations.

The combustion variables of the spherical and cylindrical flames vary in the radial direction only. Therefore, the local coordinate $s$ in the flamelet equations (cf. Eqs. (2.12), (2.13), and (2.14)) is directly related to the radial coordinate $r$. As mentioned before, the area field $\sigma$ becomes $\sigma = r^2$ for the spherical flames and $\sigma = r$ for the cylindrical flames. Furthermore, for these spherical and cylindrical flames, a relatively simple expression for the flame stretch field $K$ can be derived from the expression based on the mass-based definition (1.11):

$$K_M = \frac{\nu_f}{\sigma} \frac{\partial \sigma}{\partial r} + \frac{\partial \nu_f}{\partial r} ,$$  \hspace{2cm} (2.15)

in which the term $(1/\rho) d\rho/dt$ is not present, because the density is chosen as the progress variable, which causes this term to give no contribution to the flame stretch rate. The first term on the right-hand side of Eq. (2.15) represents the contribution to flame stretch due to the change of curvature of a moving flame surface. This term becomes $2\nu_f / r$ for spherical flames and $\nu_f / r$ for cylindrical flames. The last term of Eq. (2.15) accounts for flame thickness variations due to different flame propagation velocities of different flame contours. It appears due to the fact that the thickness of the flame is taken into account in the mass-based definition. This term does, therefore, not appear in the expression based on the area-based definition (1.4):

$$K_A = \frac{\nu_f}{\sigma} \frac{\partial \sigma}{\partial r} .$$  \hspace{2cm} (2.16)

2.3 Strategy and implementation

In Section 2.3.1, the strategy is explained to solve the three parts of the flamelet model that is presented in the previous section. The numerical implementation of the differential equations is discussed in Section 2.3.2.
2.3.1 Computational strategy

The flame-adapted coordinate system as described in Section 2.2.3 is used to solve the three parts of the flamelet model numerically. For the spherical and cylindrical flames, the coordinate perpendicular to the flame surface \( s \) and the radial coordinate \( r \) are directly related to each other: \( dr = ds \). Therefore, Eqs. (2.4), (2.5), (2.15), and (2.16) can be written in terms of the \( s \)-coordinate instead of the \( r \)-coordinate:

\[
\frac{\partial \rho}{\partial t} + v_r \frac{\partial \rho}{\partial s} = 0 ,
\]

(2.17)

\[
\frac{1}{\sigma} \frac{\partial}{\partial s} (\sigma v'_r) = \frac{s_i n'_f \frac{\partial \rho}{\rho}}{\frac{\partial s}{\partial s}} ,
\]

(2.18)

\[
K_M = \frac{v'_r}{\sigma} \frac{\partial \sigma}{\partial s} + \frac{\partial v'_r}{\partial s} ,
\]

(2.19)

\[
K_A = \frac{v'_r}{\sigma} \frac{\partial \sigma}{\partial s} ,
\]

(2.20)

respectively. The computational domain is one-dimensional with the flame-adapted \( s \)-coordinate ranging from -20 to 150 mm. For the spherical and cylindrical flames, the area field \( \sigma \) is related to the \( s \)-coordinate as:

\[
\sigma = \begin{cases} 
  (-n'_f s + C_r) & \text{if } -n'_f s + C_r > 10^{-3} \\
  10^{-3} & \text{if } -n'_f s + C_r \leq 10^{-3} 
\end{cases}
\]

(2.21)

where \( i = 1 \) for cylindrical flames and \( i = 2 \) for spherical flames. Furthermore, \( C_r \) is a constant that is equal to the radius of the flame at \( s = 0 \) and \( n'_f \) can be obtained from Eq. (2.7) with \( \partial \rho/\partial s \) instead of \( \partial \rho/\partial r \). The area field \( \sigma \) is not allowed to become smaller than \( 10^{-3} \), because of the loss of accuracy in the case that Eqs. (2.12), (2.13), (2.14), (2.18), (2.19), and (2.20) would be solved using small area fields. Notice that \( \sigma = 10^{-3} \) corresponds to a radius of one millimeter for both spherical and cylindrical flames.

The computational strategy is shown schematically in Figure 2.2 and is explained in more detail here. This strategy is based on the main idea behind the flamelet description that the time scales of the internal flame structure are much smaller than that of the movement of the flame itself, which indicates that the flame structure is changed only slightly while moving. Therefore, the entire flame structure is moved by varying the area field \( \sigma \) with a relatively large step. Subsequently, the flame structure adapts itself to the new area field by stabilizing the flame on the computational domain with the flame-adapted \( s \)-coordinate.

In order to start the computations, initial estimations are required for the flame stretch field \( K \) and the area field \( \sigma \). The flamelet equations (2.12), (2.13), and (2.14) are solved using these initial fields and using estimations for the inner structure of the flame in terms of the density \( \rho \), the mass fractions \( Y_i \), and the enthalpy \( h \). The inner structure of a steady flat stretchless flame can be used for this
2.3 Strategy and Implementation

Figure 2.2: The computational strategy.

Purpose. In these flamelet computations, the flame is fixed at \( s = 0 \) where the temperature has a certain value, which is chosen to be 500 K. From the flamelet equations, the density, the mass fractions, the enthalpy, and the mass burning rate are obtained as a function of the \( s \)-coordinate. Notice that the mass burning rate follows from the flamelet equations as an eigenvalue. Subsequently, the mass burning rate and the density are used to solve Eqs. (2.6) and (2.18) to obtain the flame propagation velocity and gas velocity throughout the computational domain. In order to solve Eq. (2.18) the gas velocity is assumed to be zero at the position where the area field is the smallest: \( \sigma = 10^{-3i} \). This assumption is justified, because symmetry considerations ensure that the gas velocity is zero at the centre of the spherical or cylindrical flames where the area field \( \sigma \) is zero. Using the resulting flame propagation velocity, the density is computed from the kinematic equation (2.17). The density obtained from this kinematic equation \( \rho^* \) and the density obtained from the flamelet equations \( \rho \) are compared with each other. In the case that these two densities are identical, the area and stretch fields are
correct and corresponding to the other combustion variables. The densities are considered to be equal to each other if the following criterion is fulfilled:

$$\left| \frac{\rho - \rho^*}{\rho^*} \right| < 10^{-5}. \tag{2.22}$$

In the case that the densities are not equal, a new estimate for the flame stretch rate has to be made by inserting the flame propagating velocity into Eq. (2.19) or (2.20). The above procedure is repeated with a new estimation of the flame stretch rate until the densities satisfy (2.22).

As soon as the criterion (2.22) is satisfied, the area field $\sigma$ is slightly changed by adding a constant $C_0$ to the constant $C_r$ in Eq. (2.21): $C_r \rightarrow C_r + C_0$. In this chapter, the computations start with $C_r = 5.0 \text{ mm}$ and this value is increased with steps of $0.25 \text{ mm}$: $C_0 = 0.25 \text{ mm}$. With the new area field obtained from Eq. (2.21), a new estimation of the flame stretch rate can be computed from Eq. (2.19) or (2.20). The change of the area field is assumed to change the inner structure of the flame only slightly. Therefore, the density, the mass fractions, and the enthalpy are good estimates for the flamelet computations with the new area field and flame stretch rate. The procedure shown in Fig. 2.2 and described above is repeated until the constant $C_r$ has reached its desired value.

Notice that, in order to solve the kinematic equation (2.17), the increment in time has to be known. This time step can be obtained from $v_{f}^r = dr/dt$ at $s = 0$, in which $dr = C_0 = 0.25 \text{ mm}$ and $v_{f}^r$ is the averaged flame propagation velocity computed from the flame propagation velocities at two successive area fields. It is also possible to obtain this time step from another position in the flame. In that case, the density iso-surface at this position $s$ in the flame can have another position $s$ after the time step. To obtain $dr$ the variation of $s$ has to be added to $C_0$ and the averaged flame propagation velocity has to be obtained from these positions in the flame. It is recognized that the computation of the time step induces an error, because the flame propagation velocity does not change linearly between two successive area fields. This error reduces for smaller constants $C_0$ and it was shown that the error is very small for $C_0 = 0.25 \text{ mm}$.

As mentioned in the previous section, the area field can be expressed in terms of the radius of the sphere or cylinder: $\sigma = r^2$ for spherical flames and $\sigma = r$ for cylindrical flames. From this knowledge the combustion variables can be expressed as a function of the radius of the sphere or cylinder. The velocities, the flame stretch rates, and the flame thickness variations obtained from the flamelet computations are shown and analysed in the next sections.

As discussed in Section 2.1, the main advantage of the flamelet description is the large reduction of computational costs without losing significant accuracy. The reduction of the computational costs turned out to be at least a factor of hundred compared to direct numerical simulations of the instationary combustion equations. Such a large reduction can also be estimated from the following simple calculation for a spherically expanding flame. The maximum time step in the instationary computations is shown to be $10^{-7} \text{ s}$. The time step in the flamelet computations is about $10^{-4} \text{ s}$, which can be computed from $v_{f}^r = dr/dt$, with a
flame propagation velocity $v_f$ of 2.5 m/s and a step in space of 0.25 mm which is equal to the constant $C_0$. Furthermore, a time step in the instationary computations is about ten times faster than the time it takes to obtain a new area field in the flamelet model. Therefore, for this example, a reduction of the computational time is predicted of a factor of 100.

2.3.2 Numerical implementation

The non-linear partial differential equations (2.12), (2.13), (2.14), and (2.18) are solved numerically using a finite-volume approach to discretise the spatial part of the equations [103, 107]. This approach is second-order accurate and fully implicit. Furthermore, an adaptive non-uniform grid is used [103]. Such a grid is very useful in combustion applications, because the essential chemical processes occur in a very thin layer compared to the length of the computational domain. More details about the numerical solver can be found in [42].

2.4 Conditions

For the simulations with the flamelet model, the combustible gas mixture that is used in the remainder of this chapter consists of methane and air with an equivalence ratio of 1.0 ($\phi = 1.0$). Besides these stoichiometric flames, the flamelet model can also be used to compute lean ($\phi < 1.0$) and rich ($\phi > 1.0$) flames. However, the results of these flames are not shown in this chapter, because, although the values for the velocities, flame stretch rate, and flame thickness are different, the behaviour of these variables is similar. The initial pressure is 1.0 atm and the temperature of the unburnt gases is 300 K. Furthermore, the velocities directed outward of the spherical and cylindrical flames are considered to be positive.

The flame has a finite thickness in which different layers can be distinguished. The inner layer is very small compared to the other layers in the flame. The position of the inner layer is chosen at the position in the flame where the heat release reaches its maximum (cf. Section 1.2.1).

In order to take the details of the thermo-chemical structure of the flame into account, a skeletal reaction mechanism [102] is used. This mechanism contains 25 reactions and 16 species, which is a minimal subset of the complete mechanism for methane-air flames. This subset is chosen, because a smaller amount of species and reactions imply less computational costs. Furthermore, the combustion variables of interest, namely the flame propagation velocity, mass burning rate, and flame stretch rate are predicted within two percent accuracy compared to the results obtained by using the more detailed reaction mechanism called GRI-mech 2.11 [14], which contains 175 reactions and 49 species. Also, the behaviour of these variables is identical. For spherically expanding flames, the results obtained from the skeletal mechanism and GRI-mech 2.11 are compared in Fig. 2.3. Figs. 2.3a and b show the flame propagation velocity and the mass burning rate
Figure 2.3: Comparison between the skeletal mechanism (solid lines) and GRI-mech 2.11 (dashed lines) for spherically expanding flames in terms of the flame propagation velocity (a) and the mass burning rate (b) at the inner layer as a function of the radius at the inner layer.

at the inner layer as a function of the radius, respectively.

The radii of the computed expanding and imploding flames lie between 5 and 60 mm. These limits are chosen, because they resemble the limits in a combustion vessel, which is the most often used experimental device to study spherically expanding flames (cf. [5, 44, 48]). A flame in such a combustion vessel is initiated by a central spark discharge (cf. [64]), which triggers the development of a flame kernel which is still influenced by the ignition phenomena (cf. [51]). This influence vanishes after the flame reaches a radius of about 5 mm [17]. These ignition processes and their impact on flame development are beyond the scope of this thesis. When the flame reaches a certain radius (60 mm in a combustion vessel with a diameter of 380 mm [48]), the flame develops instabilities on the flame surface and the pressure rises significantly. Below this critical radius, the pressure raise can be neglected.

2.5 Velocities

In Section 2.5.1, the mass burning rate of steady spherical and cylindrical flames is studied in order to point out the importance of the inner layer. Section 2.5.2 discusses the burning, flame propagation, and gas velocities of expanding and imploding flames.

2.5.1 Steady flames

For the steady spherical and cylindrical flames, the burning and gas velocities have the same magnitude but are pointing in opposite directions. The flame propagation velocity and the flame stretch rate are zero, because the flames themselves
Figure 2.4: The mass burning rate at different isotherms of steady spherical (a) and cylindrical (b) flames. At the centre of the flame, burnt gases are present. Solid lines: inner layer, dashed lines: $T = 305$ K, dotted lines: $T = 600$ K, dash-dotted lines: $T = 1900$ K, dot-dash-dotted lines: $T = 2000$ K.

do not move. For a series of computations of flames with different radii and burnt gases at the centre of the flames, the mass burning rate is plotted for the isotherms of 305, 600, 1900, and 2000K and at the inner layer (at ±1640K) in Fig. 2.4. Notice that the mass burning rate is directly related to the burning velocity by $m := \rho_s L$ (cf. Section 1.2.2).

The mass burning rate varies with the radius of the flame at all isotherms except at the inner layer. At the other isotherms almost no reactions take place, while convective and diffusive processes play the most important role. Due to the variation of the surface through which the gases flow, the gas and burning velocities and, therefore, the mass burning rate vary. This variation of the surface is obviously more dependent on the radius for spherical flames than for cylindrical flames (cf. Fig. 2.4).

At the inner layer the mass burning rate hardly varies, because here the reactions play the most important role. This can be clarified with the help of the following theoretical expression for the stretchless mass burning rate at the inner layer $m_{il}^0$:

$$
m_{il}^0 \approx \frac{1}{|Y_b^0 - Y_u|} \sqrt{\int_{Y_u}^{Y_b^0} \left( \frac{\sigma}{\sigma_{il}} \right)^2 \frac{\lambda}{L r_c p} \rho_y \, dY}, \quad (2.23)
$$

where the subscript $il$ refers to the inner layer and the superscript 0 to an unstretched flame. The derivation of Eq. (2.23) can be found in Appendix A. In the case of an infinitely thin inner layer, the integral in Eq. (2.23) effectively runs only over the infinitely thin inner layer where $\rho_y$ is a spatial delta function. This causes the term $(\sigma/\sigma_i)^2$ to become unity in Eq. (2.23), because $\sigma = \sigma_i$. No assumption is made regarding the flat, cylindrical, or spherical shape of the flame, which means that the mass burning rate is equal in all of these cases independent of the radius of the flame at the inner layer. The small deviation from the con-
stant mass burning rate in Fig. 2.4, especially for small radii, is due to the finite thickness of the inner layer. The larger the radius of the flame, the smaller the deviation, because the ratio $\sigma / \sigma_l$ approaches one.

From these notions, it can be concluded that the mass burning rate of spherical and cylindrical flames is best defined at the inner layer instead of defined at other positions in the flame. Also, in this thesis, the inner layer is considered to be the most appropriate position in the flame to evaluate the other combustion variables, such as the flame propagation velocity and the flame stretch rate.

### 2.5.2 Expanding and imploding flames

First, the burning, flame propagation, and gas velocities throughout expanding and imploding flames are studied and, subsequently, the flame propagation velocity at the inner layer is analysed.

#### Velocities throughout the flames

The burning, flame propagation, and gas velocities throughout expanding and imploding flames at a certain point in time are plotted as a function of the local radius in Figs. 2.5 and 2.6, respectively. The inner layer is located at a flame radius of 10.0 mm where the burning and gas velocities vary considerably due to the density variation through the flame. This density variation does not influence the flame propagation velocity. The small and hardly visible variation of this velocity through the flame is due to the variation in stretch. The flame propagation velocity slightly decreases in expanding flames going from the burnt to the unburnt side of the flame, which means that the flame becomes thinner at larger radii. The opposite holds for imploding flames. This effect on the flame thickness is discussed in more detail in Section 2.7.

![Figure 2.5](image)

*Figure 2.5: The burning velocity $s_L (\mbox{solid lines})$, flame propagation velocity $v_f (\mbox{dashed lines})$, and gas velocity $v'(\mbox{dotted lines})$ throughout a spherically (a) and cylindrically (b) expanding flame. The inner layer is located at a flame radius of 10.0 mm.*
2.5 Velocities

Figure 2.6: The burning velocity \( s_L n_f \) (solid lines), flame propagation velocity \( v_f \) (dashed lines), and gas velocity \( v_r \) (dotted lines) throughout a spherically (a) and cylindrically (b) imploding flame. The inner layer is located at a flame radius of 10.0 mm.

If the flame stretch expression (2.16) based on an infinitely thin flame sheet is used in the flamelet equations instead of the mass-based expression (2.15), the flame propagation velocity will become constant throughout the flame since \( \partial v_f / \partial r = 0 \) in this expression. This implies that the flame thickness and the inner flame structure do not change with varying curvature of the flame, which is obviously inconsistent with reality.

Figure 2.7: The comparison between asymptotic results (solid lines) and numerical results (dashed line) of the burning velocity throughout a spherically expanding flame.

Figs. 2.5 and 2.6 show that the absolute value of the burning velocity is higher at the burnt side of the flame than at the unburnt side and varies with the radial coordinate at the unburnt side. This behaviour can be analytically approximated using an asymptotic analysis. Here, this asymptotic analysis is performed for expanding flames. The flame propagation velocity is assumed to be uniform throughout the flame. Furthermore, it is assumed that the density jumps from its
unburnt $\rho_u$ to its burnt $\rho_b$ value at the inner layer. The gas velocity of the burnt gases is zero, because these gases are located at the centre of the sphere or cylinder. Therefore, the burning velocity at the burnt side of the flame is equal to the flame propagation velocity (cf. Eq. (2.6)):  

\[ (s_{L n_f})_b = v_f^r. \]  

(2.24)

At the inner layer, the relation $\rho_u(s_{L n_f})_l = \rho_b(s_{L n_f})_b$ can be deduced from Eq. (2.12). At the unburnt side, no reactions take place, indicating that the variation of the gas velocity is due to the variation in area through which the gases flow $\sigma v^r = (\sigma v^r)_l$. Combining these two relations and using Eqs. (2.6) and (2.24), the analytical expression for the burning velocity at the unburnt side of the flame becomes:

\[ (s_{L n_f})_u = v_f^r \left(1 - \frac{\sigma b}{\sigma u} \frac{\rho_b - \rho_u}{\rho_u}\right). \]  

(2.25)

The constant value for the flame propagation velocity in Eqs. (2.24) and (2.25) can be obtained from the numerical computations performed with the area-based expression for the flame stretch (2.16). In Fig. 2.7, the analytical results from Eqs. (2.24) and (2.25) and the numerical results from Fig. 2.5a are shown for the burning velocity of a spherically expanding flame. The behaviour of the burning velocity is well captured at the unburnt and burnt side of the flame. The small quantitative disagreement is due to the approximations made in the derivation of the analytical expressions.

Flame propagation velocity at the inner layer

For a series of computations at different flame radii, the flame propagation velocity at the inner layer is plotted as a function of the radius at the inner layer in Fig. 2.8. The flame propagation velocity shows a large dependence on the radius especially at small radii.

In Fig. 2.8, the flamelet computations are compared to direct numerical simulations of the expanding and imploding flames that are spherical and cylindrical. The direct numerical simulations were performed by solving the instantaneous combustion equations (cf. [42] for information about the numerical solver). This numerical approach is known to give very accurate results and to be computationally very expensive. Fig. 2.8 shows that the flame propagation velocities obtained from these two numerical methods agree very well. The difference is hardly visible. The flamelet computations of the spherically expanding flame is also compared to experimental data from Gu et al. [47] (cf. Fig. 2.8a). Gu et al. follow the flame at the isotherm of 305K. However, due to the very small variation of the propagation velocity through the flame, the difference between the flame propagation velocities at different isotherms is very small (cf. Figs. 2.5 and 2.6). The numerical and experimental data agree very well, except for the periodic oscillations in the experimental data. According to Gu et al., these oscillations
Figs. 2.4, 2.5, 2.6, and 2.8 show that the mass burning rate and velocities of spherical flames depend stronger on the radius than the mass burning rate and velocities of cylindrical flames. This is due to the larger curvature effects on spherical flames. Furthermore, the expanding and imploding flames differ in the direction and magnitude of the flame propagation velocity. The flame propagation velocity of expanding flames is about six times larger, because the burning and gas velocities are pointing in the same direction in contrast to imploding flames.

### 2.6 Flame stretch rate

The instantaneous flame stretch rates throughout an expanding and an imploding flame (cf. Eq. (2.15)) at a certain point in time are plotted as a function of the local radius in Fig. 2.9. In this figure, the inner layer is located at a radius of 10.0 mm. The flame stretch rate is not uniform throughout the flame, but varies with the local radius of the flame. Its absolute value is larger at smaller radii where the flame is more curved and, therefore, experiences more flame stretch.

The flame stretch rates are computed using Eq. (2.15) at a series of flame radii, or time steps. In Fig. 2.10, the flame stretch rates at the inner layer of each computation are plotted as a function of the radius at this layer. Flames with small radii at the inner layer experience more flame stretch than the ones with larger
The flame stretch rate throughout expanding (a) and imploding (b) flames. The inner layer is located at a flame radius of 10.0 mm. Solid lines: spherical flames, dashed lines: cylindrical flames.

radii, because these flames are more curved and, therefore, more stretched or compressed.

In Fig. 2.11, the flame propagation velocity is plotted as a function of the flame stretch rate at the isotherm of 305K and at the inner layer. It is important to notice that the relationship between the flame propagation velocity and the flame stretch rate is dependent on the isotherm at which it is plotted. This is mainly due to the variation of the flame stretch rate throughout flames as shown in Fig. 2.9. The numerical data are compared with both numerical and experimental data from Gu et al. [47]. They used a four-step reduced mechanism for their computations. Figure 2.11 shows that the experimental data agree very well with the numerical data obtained from the flamelet model.

The flame stretch rate of imploding flames is negative, indicating that the
2.6 Flame stretch rate

Figure 2.11: The flame propagation velocity as a function of the flame stretch rate at an isotherm of 305 K (solid line) and at the inner layer (dashed line) of a spherically expanding flame computed with the flamelet model. The numerical data are compared with numerical data (dotted line) and experimental data (plus signs) from [47].

Figure 2.12: The term $\frac{\partial v_f}{\partial r} \left( r_{il} \right)$ in Eq. (2.15) at the inner layer of expanding (a) and imploding (b) flames. Solid lines: spherical flames, dashed lines: cylindrical flames.

The flame is compressed instead of stretched, as is the case in expanding flames. Furthermore, the magnitude of the flame stretch is much higher for expanding than for imploding flames due to the large difference in the flame propagation velocity.

Also, spherical flames are more curved than cylindrical flames at the same radius, which explains the larger flame stretch rate of spherical flames. The flame stretch rates of the spherical flames are about twice as large as the cylindrical flames at the same radius. The term $\frac{\partial v_f}{\partial r}$ in the expression for the flame stretch rate (2.15) causes the difference not to be exactly a factor of two.

The difference between the expression for the flame stretch based on an infinitely thin flame sheet in Eq. (2.16) and the expression based on the mass of an infinitely small flame volume in Eq. (2.15) is the term $\frac{\partial v_f}{\partial r}$. This extra term is
quantified in Fig. 2.12 for both expanding and imploding flames. It is negative in both cases, which means that the absolute value of the predicted stretch rate with Eq. (2.15) is smaller than using Eq. (2.16) for expanding flames and larger for imploding flames. The difference in stretch rate is in all cases only a few percent or less which justifies the fact that this term is commonly neglected. However, this term has to be taken into account if one is interested in predicting the flame thickness variations. The negative values of the term $\partial v_r / \partial r$ agree with the results of the flame thickness behaviour, which is discussed in the next section.

2.7 Flame thickness

The thickness of steady, expanding, and imploding flames is computed using the geometrical definition in Eq. (1.12) and the definition of de Goey and ten Thije Boonkkamp in Eq. (1.15). In the last-mentioned definition, $s_b$ was chosen at the position in the burnt side of the flame where the heat release is three times smaller than its maximum. The results are plotted as a function of the radius of the flame at the inner layer in Figs. 2.13 and 2.14. The flame thickness resulting from the definition of Göttgens et al. in Eq. (1.13) is not computed, because the burning velocity at the unburnt side of the flame is not well defined for spherical and cylindrical flames.

![Graphs showing flame thickness](image)

Figure 2.13: The flame thickness of spherical (a) and cylindrical (b) flames computed with Eq. (1.12). Solid lines: expanding flames, dashed lines: imploding flames, dotted lines: steady flames with burnt gases at the centre, dash-dotted lines: steady flames with unburnt gases at the centre.

The behaviour of the flame thickness is similar in Figs. 2.13 and 2.14, although its magnitude is different. By varying the position $s_b$ in Eq. (1.15), the agreement can be further improved. The dependence of the flame thickness on the flame radius, however, is considered to be more interesting here than its magnitude.

Figs. 2.13 and 2.14 show that all flames approach the same limit for the flame thickness at large radii. This limit is the flame thickness of a flat stretchless flame.
2.7 Flame thickness

The thickness of both steady and unsteady flames depends on the radius of the flame. This is mainly due to the variation of the surface through which the gases flow.

The steady spherical and cylindrical flames \((K = 0)\) are thicker than the flat stretchless flame in case burnt gases are present at the centre of the sphere or cylinder. However, these flames are thinner in case unburnt gases are present at the centre. The explanation for this flame thickness behaviour is obtained by examining the temperature profiles through the flat and curved flames, because the definitions of the flame thickness in Eqs. (1.12) and (1.15) depend on these temperature profiles. The temperature as a function of the position in the flame can be deduced from the combination of the enthalpy equation (2.13) and the expression (1.26) for the enthalpy:

\[
\frac{\partial}{\partial s} (\sigma m c_p T) - \frac{\partial}{\partial s} \left( \sigma \lambda \frac{\partial T}{\partial s} \right) = 0. \tag{2.26}
\]

In the derivation of this equation, it is assumed that the Lewis numbers of all species are equal to one: \(Le_i = 1\), and that the heat conductivity \(\lambda\) and the heat capacity at constant pressure \(c_p\) have constant values. Eq. (2.26) can be solved analytically by integrating twice and using that the product of the area field \(\sigma\) and the mass burning rate \(m\) has a constant value through the flame (cf. Eq. (2.12) with \(K = 0\) for steady flames):

\[
T(s) = T_u + (T_{il} - T_u) \exp \left( \frac{m c_p}{\lambda} \int_0^s \frac{\sigma \dot{m}}{\sigma} \, ds \right). \tag{2.27}
\]

The temperature is assumed to vary between the unburnt temperature \(T_u\) and the inner layer temperature \(T_{il}\), which is located at \(s = 0\).
The mass burning rate at the inner layer $m_{il}$ is independent of the radius (cf. Fig. 2.4). Therefore, the area $\sigma$ through which transport takes place is the only variable in Eq. (2.27) that is dependent on the shape of the flame: $\sigma = 1$, $\sigma = s + r_{il}$, and $\sigma = (s + r_{il})^2$ for flat, cylindrical, and spherical flames, respectively. In the preheat zone, $\sigma > \sigma_d$ for the flames with burnt gases at the centre. Therefore, the temperature profiles of these flames vary over a larger distance than the temperature profile of the flat stretchless flame $\sigma = \sigma_d$. This indicates that the flame with burnt gases at the centre is thicker than the flat flame (cf. Eqs. (1.12) and (1.15)). The opposite holds for the flames with unburnt gases at the centre ($\sigma < \sigma_d$). The temperature profiles of these flames vary over a shorter distance. Therefore, these flames are thinner than the flat flame. Smaller radii of the sphere or cylinder will cause its thickness to deviate more from the thickness of the flat stretchless flame.

![Figure 2.15](image.png)

**Figure 2.15:** The temperature throughout a flat stretchless flame (solid line) and throughout two steady spherical flames with burnt gases (dashed line) and unburnt gases (dotted line) at the centre of the sphere ($r_{il} = 5\, \text{mm}$). The temperature profiles were computed with Eq. (2.27).

The differences in temperature profiles between the flat and curved flames are illustrated in Fig. 2.15. The temperature profiles are obtained from Eq. (2.27) for a flat stretchless flame and two steady spherical flames with unburnt and burnt gases at the centre. The radius of the inner layer is 5mm, the unburnt and inner layer temperatures are 300K and 1640K, the mass burning rate of the inner layer is 0.42kg/m$^2$s, and the ratio of the heat conductivity and the heat capacity at constant pressure is $8.5 \times 10^{-5}$ kg/ms (cf. Eq. (1.36) with $T = T_{il}$). Fig. 2.15 shows that the temperature of the spherical flame with unburnt (burnt) gases at the centre varies over a shorter (larger) distance than the temperature of the flat stretchless flame.

The temperature profiles of the unsteady flames not only depend on the area field $\sigma$, but also on the flame stretch rate $K$. Expanding and imploding flames have burnt and unburnt gases at the centre, respectively. The flame stretch rates cause an even stronger dependence of the flame thickness on the radius of the expanding and imploding flames.
Both expanding and imploding ames get thinner while moving. However, the absolute value of the stretch rate of expanding ames decreases in that case, whereas the absolute value of the stretch rate of imploding ames increases. This implies that a higher compression of imploding ames produces thinner ames, whereas increased stretching of expanding ames produces thicker ames. The expanding ames show a larger dependence on the radius than the imploding ames due to the larger flame propagation velocities and, therefore, larger flame stretch rates of expanding ames. Also, the unsteady spherical ames show a larger dependence on the radius than the unsteady cylindrical ames, because the spherical ames endure more flame stretch at the same flame radius.

2.8 Conclusions

In this chapter, a new flamelet model was developed and evaluated. This model is especially suited for the simulation of perfectly spherical and cylindrical ames. It was proven that this flamelet model produces quantitatively accurate results of the inner structure of the flame and the flow processes. Furthermore, it was shown that this model reduces the computational costs significantly in comparison with direct numerical computations. Several parts of this flamelet model are essentially different from the flamelet models that already exist. An important difference is the use of a kinematic equation of the progress variable to describe the movement of the flame instead of using the $G$-equation. This kinematic equation allows the flame to have a finite thickness instead of regarding the flame as infinitely thin. This equation is also used in combination with the continuity equation to obtain the flow field. Furthermore, for the flame stretch rate, an expression is used in which the variation of the flame thickness due to the movement of the flame is taken into account. This term is small and usually neglected in other models. If one is interested in the dynamics of the flame however, it is of importance. Besides the development of this new flamelet model, a new computational strategy was developed to overcome the problem of the non-local manner with which the inner flame structure and the flow are coupled. In this strategy, the area field of the flame is varied with a relatively large step and, subsequently, the flame is stabilized on the computational domain.

With the results obtained from the computations with this new flamelet model, the behaviour of various combustion variables was analysed and several interesting aspects were discovered. One of these new insights is that, at the inner layer, the mass burning rate of steady spherical and cylindrical ames is almost independent of the flame radius. For ames with an infinitely thin inner layer, the mass burning rate appeared to be even completely independent of the flame radius. These statements were demonstrated both analytically and numerically. This independence is due to the fact that at the inner layer the chemical reactions play the dominating role rather than the convective and diffusive processes, which are more important in the preheat zone and oxidation layer. These arguments are also the main reason to define the inner layer as the most appropriate
position in the flame to evaluate the combustion variables, whereas, in literature, the unburnt or the burnt side of the flame is commonly used for this purpose. The position to evaluate the combustion variables is important, because crucial variables such as the burning velocity can vary significantly through the flame. Also, the flame stretch rate is dependent on the position in the flame. Another aspect that was found is that the thickness of the flame changes due to expansion or implosion of the spherical and cylindrical flames. These flames get thinner while moving. In literature, stretch is believed to cause flames to get thinner and compression is believed to have the opposite effect. However, it was shown here that the imploding flames also get thinner while they are compressed.
Influence of flame stretch on the mass burning rate

3.1 Introduction

Flame stretch can change the mass burning rate of premixed flames significantly. It is important to know the relation between these two parameters, because the mass burning rate is a crucial parameter in premixed combustion processes. In this chapter, the change of the mass burning rate due to flame stretch is investigated with regard to the unstretched mass burning rate. The emphasis lies on deriving expressions for the proportionality parameter that appears in the relation between the mass burning rate and the flame stretch rate. This proportionality parameter denotes the sensitivity of a flame to flame stretch and is called the Markstein number.

In Section 3.2, two distinct expressions for the mass burning rate are deduced. A first formulation is deduced from asymptotic analysis and the other one from integral analysis. From the combination of these two formulae, different expressions for the Markstein number are derived in Section 3.3. According to a phenomenological law, the Markstein number can be separated into a part for the curvature of the flame and a part for the straining of the flow. This separation is analysed in Section 3.4. In Sections 3.3 and 3.4, the Markstein numbers are computed from the theoretical expressions and numerical computations for planar stretchless and stagnation flames, spherically expanding and imploding flames, and steady spherical flames.

3.2 Expressions for the mass burning rate

Section 3.2.1 presents a relation between the ratio of the stretched and unstretched mass burning rate and the flame stretch rate that was deduced using asymptotic analysis. In this relation, the Markstein number is an unknown proportionality parameter. Section 3.2.2 discusses the dependency of this Markstein number on
the position in the flame. In Section 3.2.3, an expression for the mass burning rate at the burnt flame boundary is presented. This expression was deduced using integral analysis.

### 3.2.1 Asymptotic analysis

Markstein [69] was the first to propose a phenomenological relation between the laminar burning velocity and the curvature of the flame front:

\[
\frac{s_L}{s_L^0} = 1 + \frac{L}{R_f},
\]

where \( R_f \) is the radius of curvature of the flame front and the superscript 0 refers to an unstretched flame. In this relation, the straining of the flow is not taken into account. In 1985, Clavin [26] derived a relation between the burning velocity and the flame stretch rate thereby taking both curvature of the moving flame surface and straining of the flow into account. The derivation of this relation is based on the asymptotic analysis of Clavin and Williams [25] and of Matalon and Matkowsky [70]:

\[
\frac{s_L}{s_L^0} = 1 - \frac{KL}{s_L^0} + O \left( \epsilon^2 \right),
\]

where \( \epsilon \) is the perturbation parameter, which is the ratio of the flame thickness to the characteristic wrinkle size of the flame front. In the relations (3.1) and (3.2), the proportionality of the burning velocity to the flame stretch is represented by the parameter \( L \). This parameter is considered as a characteristic length assumed to be of the order of the thickness of the flame and is generally known as the Markstein length. Instead of the Markstein length, the dimensionless Markstein number \( M \) is also often used, which is simply the Markstein length divided by the flame thickness \( M := L/\delta_f \). The flame thickness is defined here as a diffusive length scale (cf. Eq. (1.13)). Relation (3.2) can be rewritten in terms of the Markstein number, the mass burning rate \( m := \rho s_L \) (cf. Section 1.2.2), and the Karlovitz number

\[
K_a := \left( \frac{\rho \delta_f}{m} \right)^0 \mathcal{K},
\]

which represents a dimensionless flame stretch rate:

\[
\frac{m}{m^0} = 1 - K_a M + \text{hot}.
\]

The term \text{hot} indicates higher-order terms in the Karlovitz number. These higher-order terms take into account that the structure of the flame changes due to stretch, which influences the mass burning rate of flames that endure stronger stretch rates.
3.2 Expressions for the Mass Burning Rate

3.2.2 Position in the flame

The value of the Markstein number in Eq. (3.4) depends on the position in the flame at which it is evaluated, because the mass burning rate and the flame stretch rate change going from the unburnt to the burnt side of the flame. This is shown in Fig. 3.1. In this figure, the ratio between the stretched and unstretched mass burning rate is plotted as a function of the Karlovitz number for a spherically expanding flame at different isotherms with $\phi = 1.0$ and $Le_i \neq 1$. These results are obtained from the computations shown in Chapter 2. Notice that the unstretched mass burning rate is not obtained from a flat unstretched flame, but from a steady spherical flame. The Karlovitz number is computed using Eq. (3.3) in which the flame thickness is 0.202mm. This flame thickness is computed from the expression derived by Göttgens et al. in Eq. (1.13). The Markstein numbers for the different isotherms in Fig. 3.1 are obtained from the slopes of the curves that are extrapolated to zero stretch (cf. Eq. (3.4)). The corresponding Markstein numbers at 305K, 500K, 900K, the inner layer, and 1900K are -1.06, 0.02, 0.91, 2.92, and 5.31, respectively. These Markstein numbers are quite different at different isotherms and can even become negative at the unburnt side of the flame.

![Figure 3.1: The ratio of the stretched and unstretched mass burning rate as a function of the Karlovitz number at different isotherms of a spherically expanding flame, with $\phi = 1.0$ and $Le_i \neq 1$. Solid line: inner layer, dashed line: $T = 305K$, dotted line: $T = 500K$, dash-dotted line: $T = 900K$, dot-dash-dotted line: $T = 1900K$.](image)

Therefore, a choice has to be made at which isotherm the Markstein number will be evaluated in the remainder of this chapter. In other studies, the unburnt or burnt flame boundary is often chosen to evaluate the Markstein number. However, the choice is made here to evaluate the Markstein number at the inner layer, because here most of the hydrocarbon chemistry takes place. Furthermore, the mass burning rate of a stretchless flat flame differs from the mass burning rate of a steady and, therefore, unstretched cylindrical or spherical flame at the unburnt and the burnt flame boundary. However, it has been shown analytically (cf. Eq. (2.23)) and numerically (cf. Fig. 2.4) that only at the inner layer of the flame the mass burning rates of these flames are equal to each other.
3.2.3 Integral analysis

In the weak stretch limit, de Goey and ten Thije Boonkkamp [40,108] have derived the following expression for the mass burning rate at the burnt side of the flame $m_b$ in terms of the flame stretch rate in which no proportionality parameter such as the Markstein number is present:

$$
\frac{m_b}{m_b^0} = 1 - IK_T^0 + (h_b - h_u) \frac{\partial \ln (m_b^0)}{\partial h_b^0} + \sum_{j=1}^{N_e} \left( \psi_{j,b} - \psi_{j,u} \right) \frac{\partial \ln (m_b^0)}{\partial \psi_{j,b}^0} + \text{hot},
$$

(3.5)

with

$$
h_b - h_u = - \sum_{i=1}^{N_s} M_i h_i^0 \left( \frac{IK_i^0}{Le_i} - IK_T^0 \right) \left( \phi_i^{0,b} - \phi_i^{0,u} \right) + \text{hot},
$$

(3.6)

and

$$
\psi_{j,b} - \psi_{j,u} = - \sum_{i=1}^{N_s} \mu_{ji} \left( \frac{IK_i^0}{Le_i} - IK_T^0 \right) \left( \phi_i^{0,b} - \phi_i^{0,u} \right) + \text{hot}.
$$

(3.7)

In Eqs. (3.5), (3.6), and (3.7), $N_e$ denotes the number of elements, $\mu_{ji}$ the number of elements $j$ in species $i$, $\phi_i$ the mole number of species $i$ that is defined as $\phi_i := Y_i/M_i$, and $\psi_j$ the mole number of element $j$ that is defined as $\psi_j := \sum_{i=1}^{N_s} \mu_{ji} Y_i/M_i$. Furthermore, the Karlovitz integrals $IK_i^0$ and $IK_T^0$ determine the integrated effect of stretch in the flame structure between the unburnt and burnt flame boundaries:

$$
IK_i^0 := \frac{Le_i}{(\sigma m^0)_b} \int_{s_u}^{s_b} \sigma_0^0 K_0^i ds
$$

(3.8)

and

$$
IK_T^0 := \frac{1}{(\sigma m^0)_b} \int_{s_u}^{s_b} \sigma_0^0 KT^0_0 ds.
$$

(3.9)

The normalized unstretched mole number $\bar{\phi}_i^0$ and temperature field $\bar{T}_0$ in Eqs. (3.8) and (3.9) are defined by

$$
\bar{\phi}_i^0 := \frac{\phi_i^{0,b} - \phi_i^{0,u}}{\phi_i^{0,b} - \phi_i^{0,u}}
$$

(3.10)

and

$$
\bar{T}_0 := \frac{T^0 - T_u}{T_b^0 - T_u}.
$$

(3.11)

Eq. (3.5) can be derived by applying the integral analysis of Chung and Law [24] to the flamelet equations for the mass (2.12), for the enthalpy (2.13), and for
the mole number of element $j$:

$$
\frac{\partial}{\partial s} \left( \sigma m \psi_j \right) - \frac{\partial}{\partial s} \left( \sigma \frac{\lambda}{c_p} \frac{\partial \psi_j}{\partial s} \right) = -\frac{\partial}{\partial s} \left( \sigma \frac{\lambda}{c_p} \sum_{i=1}^{N} \left( \frac{1}{Le_i} - 1 \right) \frac{\mu_{ji}}{M_i} \frac{\partial Y_i}{\partial s} \right) = -\sigma \rho K \psi_j.
$$

Eq. (3.12) can be derived from the conservation equation for the mole number of element $j$:

$$
\frac{\partial}{\partial t} (\rho \psi_j) + \nabla \cdot (\rho \mathbf{v} \psi_j) = \nabla \cdot \left( \frac{\lambda}{c_p} \nabla \psi_j \right) + \nabla \cdot \left( \frac{\lambda}{c_p} \sum_{i=1}^{N} \left( \frac{1}{Le_i} - 1 \right) \frac{\mu_{ji}}{M_i} \nabla Y_i \right),
$$

in the same way as the derivation of the flamelet equations in Section 2.2.3. The details of the derivation of Eq. (3.5) can be found in [40, 108].

The physical meaning of the different terms in Eq. (3.5) is explained hereafter. Flame stretch can modify the transport of heat from the reaction layer to the preheat zone. This effect is included in the second term $IK^0 T$ on the right-hand side of Eq. (3.5). The third term $(h_b - h_{ij}) \partial \ln(m^0_b) / \partial h^0_b$ and the fourth term $\sum_{j=1}^{N} (\psi_{j,b} - \psi_{j,u}) \partial \ln(m^0_b) / \partial \psi^0_{j,b}$ denote the effect of preferential diffusion on the change in the local enthalpy and on the change in the element composition and the stoichiometry, respectively. Without preferential diffusion, only the term $1 - IK^0 T$ does not vanish. The last term hot indicates higher-order terms in the Karlovitz integrals $IK^0 T$ and $IK^0 J$. Notice that in order to obtain the mass burning rate at the burnt flame boundary from Eq. (3.5), the behaviour of the flame stretch rate throughout the flame has to be known.

### 3.3 Combined Markstein number

The goal is to derive expressions for the Markstein number at the inner layer. In Sections 3.3.1 and 3.3.2, the relation between the mass burning rate and flame stretch rate in Eq. (3.5) is used in combination with the definition of the Markstein number in Eq. (3.4) in order to obtain these theoretical expressions for the combined Markstein number. This combined Markstein number represents the sensitivity of the flame to both the curvature of the flame and the straining of the flow. In Section 3.3.1, it is assumed that the reaction layer is infinitely thin. This assumption is not made in Section 3.3.2. In Section 3.3.3, the values of the Markstein numbers obtained from the theoretical expressions are compared with each other and with numerical computations for both unit and non-unit Lewis numbers.
3.3.1 Infinitely thin reaction layer

In this section, it is assumed that the reaction layer is infinitely thin, which means that the inner layer and the burnt flame boundary have the same position in the flame. Furthermore, planar stagnation flames and spherical flames are considered separately.

**Planar stagnation flames**

For planar stagnation flames, the local flame surface area is equal to one. Furthermore, in the case of weak stretch, the flame stretch rate reduces to a uniform field throughout the flame. By combining the relations (3.4) and (3.5) and using the unstretched profiles of temperature and mole numbers in the preheat zone ($s < s_b$) together with $\rho^0(s) = \rho_u T_u / T^0_b(s)$ in the Karlovitz integrals (3.8) and (3.9), the following expression for the Markstein number at the burnt flame boundary is found:

$$M_b = \left( \frac{1 + \tau}{\tau} \right) \ln (1 + \tau) + (1 + \tau) F_{fuel} \left( L_{fuel} - 1 \right) \left( T^0_b - T_u \right) \frac{\partial \ln (m^0_b)}{\partial T^0_b},$$

with

$$F_{fuel} = \int_0^\tau \frac{\ln (1 + y)}{y^2} \left( \frac{y}{\tau} \right) L_{fuel} dy$$

and $\tau$ the thermal expansion coefficient, which is defined as

$$\tau := \frac{T^0_b - T_u}{T_u} = \frac{\rho_u - \rho^0_b}{\rho^0_b}.$$  \hspace{1cm} (3.14)

The details of the derivation of Eq. (3.14) can be found in Appendix B.

In deriving Eq. (3.14), it is assumed that $m^0_b$ depends directly on $T^0_b$ and not on $\phi_{CO_2,b}^0$, $\phi_{H_2O,b}^0$, and $\phi_{O_2,b}^0$. If this dependence of the mass burning rate on the temperature is effectively given by the relation $m^0_b \propto \exp(-E_a/2RT^0_b)$, the term $(T^0_b - T_u)\partial \ln (m^0_b)/\partial T^0_b$ in Eq. (3.14) becomes $Z_e^0/2$ with $Z_e^0$ the unstretched Zeldovich number, which is defined as

$$Z_e^0 := \frac{E_a(T^0_b - T_u)}{R(T^0_b)^2}.$$  \hspace{1cm} (3.17)

Using this relation, the expression for the Markstein number at the burnt flame boundary derived by Clavin [26] can be obtained from Eq. (3.14). Clavin used Large Activation Energy Asymptotics (LAEA) to derive an expression for the Markstein number. He assumed that $(L_{fuel} - 1)$ is of the order $1/Z_e^0$. Therefore,
to the lowest order in the Zeldovich number, Eq. (3.14) reduces to the expression for the Markstein number $M_b$ derived by Clavin [26]:

$$M_b = \left(1 + \tau \frac{1 + \tau}{\tau}\right) \ln \left(1 + \tau^2\right) + \frac{Z\varepsilon^0_0}{2\left(Le_{fuel} - 1\right)} \left(1 + \tau \frac{1 + \tau}{\tau}\right) \ln \left(1 + \frac{y}{y} \right) dy.$$  (3.18)

Peters and Williams [85] showed that the above ad-hoc relation for the mass burning rate $m_0^b = \exp\left(-E_a/2RT_b^0\right)$ should be replaced by $m_0^b = \left(T_b^0 - T_a\right)^2$. In order to find this relation, they used a more complete theory of the flame structure that is based on the systematic reduction of the detailed reaction mechanism in combination with rate-ratio asymptotics. Using this relation instead, the term $(T_b^0 - T_a)\ln(m_0^b/\partial T_b^0)$ in Eq. (3.14) becomes $2(T_b^0 - T_a)/(T_b^0 - T_d) = 2/\varepsilon\varepsilon_0$, where $\varepsilon\varepsilon_0$ is a measure for the relative thickness of the oxidation layer compared to the total flame thickness. Note that $4/\varepsilon\varepsilon_0$ can be considered as an effective Zeldovich number. If this relation is inserted in Eq. (3.14) and if $(Le_{fuel} - 1)$ is of the order $\varepsilon\varepsilon_0$, the same expression for the Markstein number as derived by Rogg and Peters [95] is obtained:

$$M_b = \left(1 + \tau \frac{1 + \tau}{\tau}\right) \ln \left(1 + \tau^2\right) + \frac{2}{\varepsilon\varepsilon_0} \left(1 + \tau \right) F_{fuel} \left(Le_{fuel} - 1\right).$$  (3.19)

### Spherical flames

For spherically expanding or imploding flames, the flame stretch rate is given by $K(r) = 2\nu_f^0(r)/(r + \partial\nu_f^0(r)/\partial r)$, where $r$ is the radial coordinate (cf. Eq. (2.15), with $r = r^2$ for spherical flames). The spherical flames are considered to be weakly stretched, which means that the radius at the burnt flame boundary $r_b$ is large compared to the flame thickness $\delta_f^0$. Therefore, the dependence of the flame propagation velocity on the radius $\nu_f(r)$ is neglected here (cf. Fig. 2.12). Furthermore, the radial coordinate is approximated by $r = r_b(1 + \mathcal{O}(\delta_f^0/r_b))$. The above assumptions reduce the Karlovitz number for spherical flames to $K_{ab} := 2\rho_b^0 \delta_f^0 \nu_f^0/m_0^b r_b$.

The undistorted profiles for $T^0(r)$ and $\phi_f^0(r)$ are the profiles of steady spherical flames, for which the flame stretch rate and the flame propagation velocity are equal to zero. For the weakly stretched spherical flames, these profiles are equal to the exponentials in Eqs. (B.12) and (B.13) with an error of $\mathcal{O}(\delta_f^0/r_b)$. The mass burning rate $m_0^b$ is given by the mass burning rate of a steady spherical flame, which is in general not equal to the mass burning rate of a stretchless flat flame. However, at the inner layer and, therefore, at the burnt flame boundary, in the case of an infinitely thin reaction layer, the mass burning rates of steady spherical and stretchless flat flames are equal (cf. Eq. (2.23) and Fig. 2.4).

Substitution of all this information into the Karlovitz integrals in Eqs. (3.8) and (3.9) gives the same results as in Eqs. (B.15) and (B.16) for the case of a planar stagnation flame, up to order $\mathcal{O}(\delta_f^0/r_b)$. This means that the same expressions
for the Markstein number in Eqs. (3.14) and (3.19) are obtained for spherically imploding and expanding flames as well as for stagnation flames in the limit of zero stretch.

### 3.3.2 Finite thickness of the reaction layer

In realistic flames, the reaction layer has a finite thickness, which indicates that the inner layer and the burnt flame boundary are not present at the same position in the flame. In order to find an expression for the Markstein number at the inner layer, the relations between the mass burning rate and the flame stretch in Eqs. (3.14) and (3.19) can be combined. The mass burning rates in Eqs. (3.14) and (3.19) are evaluated at the inner layer and the burnt flame boundary, respectively. Therefore, a relation between these different mass burning rates is required. This relation is given by the conservation equation of mass written in a flame-adapted coordinate system (2.12).

For weakly stretched flames, the flame stretch rates in the Karlovitz integrals in Eqs. (3.8) and (3.9) are assumed to be uniform throughout the flame and the local flame surface area is assumed to be equal to one. By integrating Eq. (2.12) from the inner layer to the burnt flame boundary and combining it with Eqs. (3.4) and (3.5), the expression for the Markstein number at the inner layer $M_{il}$ becomes:

$$
M_{il} = \frac{1}{\int_{s_u}^{s_b} \rho^0 \tilde{T}_b^0 \, ds - \int_{s_b}^{s_u} \rho^0 \, ds}
$$

$$
- \left( \tau_b^0 - \tau_u^0 \right) \frac{\partial \ln (m_b^0)}{\partial \tilde{T}_b^0} \int_{s_u}^{s_b} \rho^0 \left( \tilde{\phi}_{fuel}^0 - \tilde{T}_0^0 \right) \, ds.
$$

Expression (3.20) for the Markstein number $M_{il}$ is only dependent on unstretched flame data. It requires knowledge of the inner structure of an unstretched flame. The last term in Eq. (3.20) is due to preferential diffusion.

Notice that Eqs. (3.14), (3.19), and (3.20) are valid for lean hydrocarbon-air flames (cf. Appendix B). For stoichiometric flames, besides fuel, other species have to be taken into account as well. However, if preferential diffusion is not taken into account, these equations can also be used for stoichiometric flames.

### 3.3.3 Theoretical and numerical results

In this section, the Markstein numbers are computed from the theoretical expressions (3.19) and (3.20). Also the Markstein number is computed directly from Eq. (3.4) by using numerical simulations on weakly stretched flames and extrapolating these numerical results to zero stretch. Furthermore, the relation between the mass burning rate and the flame stretch rate is investigated for stronger stretch. This section ends with a discussion about the difficulty to compare the
Table 3.1: The temperature at the inner layer and the unstretched flame thickness for $\phi = 0.8$ and $\phi = 1.0$ and for both cases of unit and non-unit Lewis numbers.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>$Le_i = 1$</th>
<th>$Le_i \neq 1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_i$ [K]</td>
<td>1518 1662</td>
<td>1493 1640</td>
</tr>
<tr>
<td>$\delta_i^0$ [mm]</td>
<td>0.272 0.255</td>
<td>0.259 0.202</td>
</tr>
</tbody>
</table>

Markstein numbers computed here with the Markstein numbers obtained in other studies.

Planar stretchless and stagnation flames as well as steady, imploding, and expanding spherical flames are considered. The flamelet model described in Chapter 2 is used to compute the spherical flames. The planar flames (cf. [31, 61]) are computed from the flamelet equations (2.12), (2.13), and (2.14), in which $\sigma = 1$ and the coordinate perpendicular to the flame surface $s$ is equal to the $x$-coordinate in a cartesian coordinate system $x = (x, y, z)$. For the stretchless flames, the flame stretch rate $K$ in these flamelet equations is zero. For the stagnation flames, the flame stretch rate $K$ is obtained from a differential equation for the flame stretch rate, which is derived from the conservation equations of momentum (1.17):

$$\frac{d}{dx} \left( \mu \frac{dK}{dx} \right) = \rho_a a^2 - 2 \rho K^2, \quad (3.21)$$

with $a$ the applied strain rate, which is equal to the flame stretch rate in the unburnt mixture: $a = K (x \to -\infty)$. More details about the computations of the planar stagnation flames and about the derivation of Eq. (3.21) can be found in [79].

For the numerical computations, the skeletal reaction mechanism in [102] is used and the initial pressure is 1.0 atm. Furthermore, the gas mixture consists of methane and air, the temperature of the unburnt gases is 300 K, and the Lewis number for methane $Le_{CH_4}$ is 0.97. Both cases of unit and non-unit Lewis numbers are considered. The non-unit Lewis numbers are spatially independent. The unit Lewis numbers have been used in order to be able to compare the theoretical and numerical computations without the influence of preferential diffusion, although this model has no direct physical meaning. For the case of unit Lewis numbers, both lean ($\phi = 0.8$) and stoichiometric ($\phi = 1.0$) flames are considered and for the case of non-unit Lewis numbers, only lean ($\phi = 0.8$) flames are considered.

Values for the inner layer temperature and the flame thickness are required in the computations. The inner layer temperature is defined at the position in the flame where the heat release reaches its maximum and the flame thickness is computed from Eq. (1.13). These parameters are dependent on the equivalence ratio and the Lewis numbers. In Table 3.1, the values of these parameters are shown for the different situations considered in this chapter.
Table 3.2: Markstein numbers computed directly from the numerical computations (cf. Eq. (3.4)) and from the theoretical expressions in Eqs. (3.19) and (3.22) for $\phi = 0.8$ and $\phi = 1.0$, and for $Le_l = 1$.

<table>
<thead>
<tr>
<th>Equation</th>
<th>$\phi = 0.8$</th>
<th>$\phi = 1.0$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\tau M_{il}$</td>
<td>$\tau M_{il}$</td>
</tr>
<tr>
<td>Eq. (3.4)</td>
<td>1.12</td>
<td>1.18</td>
</tr>
<tr>
<td>Eq. (3.19)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>inner layer</td>
<td>4.06</td>
<td>2.02</td>
</tr>
<tr>
<td>burnt side</td>
<td>5.68</td>
<td>2.23</td>
</tr>
<tr>
<td>Eq. (3.22)</td>
<td>1.19</td>
<td>1.24</td>
</tr>
</tbody>
</table>

**Unit Lewis numbers**

The Lewis numbers of all species are assumed to be equal to one. Therefore, there is no preferential diffusion and the Karlovitz integrals $IK_0^0$ and $IK_0^T$ are equal to each other (cf. Eqs. (3.8) and (3.9)). This indicates that the expression for the Markstein number at the inner layer in which the reaction layer has a finite thickness becomes:

$$M_{il} = \frac{1}{\partial^0 \rho_0^0} \left( \int_{s_u}^{s_b} \rho^0 T^0 \, ds - \int_{s_u}^{s_b} \rho^0 \, ds \right),$$

(3.22)

instead of Eq. (3.20). Furthermore, only the first term in Eq. (3.19) contributes to the Markstein number.

In Table 3.2, the Markstein numbers obtained from Eqs. (3.19) and (3.22) and from numerical computations are presented for lean ($\phi = 0.8$) and stoichiometric ($\phi = 1.0$) flames. The thermal expansion coefficient $\tau$ in Eq. (3.19) is computed with the temperature or density at the inner layer and at the burnt side of the flame (cf. Eq. (3.16)). The Markstein numbers from the numerical computations are equal for both the stagnation and spherical flames. This was expected, because the combined Markstein number is uniquely defined. The theoretical and numerical predictions show that the Markstein number is lower for the lean flames than for the stoichiometric flames, which indicates that stoichiometric flames are more sensitive to flame stretch than lean flames.

Furthermore, it is shown in Table 3.2 that the Markstein number obtained from Eq. (3.19) is predicted within a factor of two compared to the Markstein number from the numerical computations. However, quantitative agreement is not expected between these Markstein numbers, because the assumptions made in the derivation of Eq. (3.19) do not hold in the numerical computations of the Markstein number (cf. [29, 30]). The theoretical prediction of the thermal expansion coefficient computed at the inner layer is slightly different from this coefficient computed at the burnt side of the flame. The Markstein number obtained from Eq. (3.22) differs only a few percent from the Markstein number obtained from numerical computations. This means that the Markstein number is predicted very accurately if the inner structure of the flame is taken into account.
3.3 Combined Markstein number

Figure 3.2: The ratio of the stretched and unstretched mass burning rate as a function of the Karlovitz number for $\phi = 0.8$ (a) and $\phi = 1.0$ (b), and for $Le_i = 1$. Solid lines: weak stretch limit from Eq. (3.22), dashed lines: spherically expanding flame, dotted lines: stagnation flame.

Figure 3.3: The first derivative of the flame stretch rate $\partial K/\partial s$ at the inner layer as a function of the Karlovitz number for $\phi = 0.8$ (a) and $\phi = 1.0$ (b), and for $Le_i = 1$. Dashed lines: spherically expanding flame, dotted lines: stagnation flame.

It is, however, more elaborate to obtain the Markstein numbers from Eq. (3.22), because the density and the temperature or methane fraction have to be known throughout an unstretched flame, whereas Eq. (3.19) only requires the unburnt and burnt values of the density.

The Markstein number derived for weakly stretched flames is less useful for stronger stretch, because the higher-order stretch terms in Eq. (3.4) play an increasing role as the flame stretch rate becomes higher. In that case, the linear relation is not valid anymore, which is illustrated in Fig. 3.2. In this figure, the ratio of the stretched and unstretched mass burning rate $m_{\text{st}}/m_{\text{uni}}$ is plotted as a function of the Karlovitz number $Ka_i$. The mass burning rates and Karlovitz
Table 3.3: Markstein numbers computed directly from the numerical computations (cf. Eq. (3.4)) and from the theoretical expressions in Eqs. (3.19) and (3.20) for $\phi = 0.8$ and $Le_i \neq 1$.

<table>
<thead>
<tr>
<th>Equation</th>
<th>$\phi = 0.8$</th>
<th>$\tau$</th>
<th>$\mathcal{M}_i$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Eq. (3.4)</td>
<td></td>
<td>2.14</td>
<td></td>
</tr>
<tr>
<td>Eq. (3.19) inner layer</td>
<td></td>
<td>3.98</td>
<td>1.39</td>
</tr>
<tr>
<td>burnt side</td>
<td></td>
<td>5.68</td>
<td>1.50</td>
</tr>
<tr>
<td>Eq. (3.20)</td>
<td></td>
<td></td>
<td>1.61</td>
</tr>
</tbody>
</table>

Figure 3.4: The ratio of the stretched and unstretched mass burning rate as a function of the Karlovitz number for $\phi = 0.8$ and $Le_i \neq 1$. Solid line: weak stretch limit from Eq. (3.20), dashed line: spherically expanding flame, dotted line: stagnation flame.

Non-unit Lewis numbers

More realistic flames with non-unit Lewis numbers were simulated numerically and computed from the theoretical expressions (3.19) and (3.20). The term $(T_0^b - T_u)\partial \ln(m_0^b)/\partial T_0^b$ in Eq. (3.20) is replaced by $2(T_0^b - T_u)/(T_0^b - T_\infty) = 2/\varepsilon_0$, as is also done in the derivation of Eq. (3.19). Furthermore, in Eq. (3.20), the temperature field $T_0^b$ has a chemical source that is broad compared to the source numbers are obtained from numerical computations. As a reference, the theoretical prediction from Eq. (3.22) for weakly stretched flames is plotted as a straight line in Fig. 3.2. This figure shows also that, for increasing stretch rates, stagnation flames deviate faster from the weak stretch limit than the spherical flames. This is due to the fact that the higher-order terms contain the variation of the stretch throughout the flame, which is larger for stagnation than for spherical flames at the same stretch rates. This is illustrated in Fig. 3.3 in which the first derivative of the flame stretch rate at the inner layer is plotted as a function of the Karlovitz number.
of the mole number for methane $\phi_{CH_4}^0$. This is due to the oxidation processes that induce the slow temperature increase in the burnt gases. These processes hardly have any influence on the preheat zone and the inner layer. Eq. (3.20) is derived for relatively thin reaction layers. In order to correct for the relatively thick reaction layer for the temperature, the burnt flame boundary in Eq. (3.20) is positioned in the flame where the mole number for methane is almost zero \(1 - \phi_{CH_4}^0 = 10^{-3}\).

The theoretical and numerical results for lean \(\phi = 0.8\) flames with more realistic Lewis numbers are shown in Table 3.3 and Fig. 3.4. The global behaviour is clearly similar to the one with unit Lewis numbers. Again, the Markstein number from the numerical computations is equal for the stagnation and spherical flames. Furthermore, Fig. 3.4 shows that the higher-order effects of the stretch on flames are larger for stagnation flames than for spherically expanding flames at the same stretch rates. The main difference between the cases of unit and non-unit Lewis numbers is that the Markstein number computed from the numerical computations is higher for the case of non-unit Lewis numbers.

Comparison with other studies

The derivation of other theoretical expressions for the Markstein number can be found in e.g. [25,95]. Also several authors have computed the Markstein number or length directly from numerical computations [17,74] or measured it [5,15,32,54,100,111].

The Markstein number strongly depends on the isotherm upon which the Markstein number is defined (cf. Fig. 3.1) and on the various definitions and expressions that are used. Therefore, it must be clearly stated which definition, isotherm, and position in the flame are used. This is the reason why it is in general difficult to compare the Markstein numbers presented here with the Markstein numbers from other numerical and experimental studies. This comparison is, therefore, not made. However, the computation of the Markstein numbers is mainly based on stretched flame data such as the burning velocities and flame propagation velocities. In Figs. 2.11 and 3.5, the numerical data of spherically expanding and stagnation flames are compared directly with experimental data from others [47,112] and show good agreement.

In Fig. 2.11, the flame propagation velocity is used to compare the data of spherically expanding flames, because this velocity does not change significantly throughout the flame as is the case for the burning velocity. Therefore, the flame propagation velocity is less dependent on the position in the flame. Stagnation flames are not moving. Therefore, the burning velocity is used to compare the data. The burning velocity in Fig. 3.5 was obtained at the same position in the unburnt gases as in the experimental study [112]. However, the numerical computations were performed with potential flow boundary conditions and the experiments with plug flow. As indicated in [29], these different inflow boundary conditions can generate different burning velocities and stretch rates at the unburnt side of the flame. It is expected that this difference is small here, because
Figure 3.5: The scaled burning velocities of stagnation flames as a function of the strain rates for $\phi = 1.0$ and $Le_i \neq 1$. Solid line: numerical data at the unburnt side, plus signs: experimental data at the unburnt side from [112].

the distance between the nozzle outlet and the flame is relatively large. Notice that the burning velocity at the unburnt side of the flame is larger for larger strain rates, whereas the burning velocity at the inner layer is smaller for larger strain rates. This indicates that the Markstein number computed at the unburnt side of the flame can be negative, which emphasizes the importance of the choice of the position in the flame at which the Markstein number is computed.

3.4 Markstein numbers for curvature and strain

Flame stretch consists of strain and curvature. These effects can be taken into account separately, thereby introducing a Markstein number for the curvature part of the flame and another one for the flow straining through the flame. In Section 3.4.1, the separation into two different Markstein numbers is discussed in more detail. Expressions for the Markstein numbers are deduced in Sections 3.4.2 and 3.4.3 for flames with an infinitely thin reaction layer and a finite thickness of the reaction layer, respectively. The theoretical and numerical results of the Markstein numbers are discussed in Section 3.4.4.

3.4.1 Concept of separate Markstein numbers

For a general flame that is weakly curved and strained, the total flame stretch rate can be written as:

$$K = K_c' + K_s' = \sum_{i=1}^{2} \frac{v_f n_i}{r_i} + \sum_{i=1}^{2} a_i,$$

(3.23)

with $a_i$ the strain rates, $r_i$ the radii of curvature related to the principal axes in the plane of the flame, and $v_f n_i = v_f \cdot n_i$ the flame propagation velocity normal to the flame surface. Eq. (3.23) can be obtained from Eq. (1.11), in which the third
term on the right-hand side is neglected, because the variation of the flame propagation velocity due to flame thickness variations is small for weakly stretched flames, and in which the last term is zero, because the density is chosen as the progress variable. The total flame stretch rate in Eq. (3.23) is a linear combination of the flame stretch rate of a moving spherical flame \( K'_c = \Sigma_{i=1}^{2} v_i^f / r_i \) and of a steady planar stagnation flame \( K'_s = \Sigma_{i=1}^{2} a_i \) (cf. Fig. 1.3 and Section 1.2.3).

The combination of the Karlovitz number and the Markstein number for the total flame stretch in Eq. (3.4) can be written as a linear combination of the curvature and flow straining part of the flame:

\[
\frac{m}{m_0} = 1 - Ka'_c M'_c - Ka'_s M'_s + \text{hot},
\]

with the subscripts \( c \) and \( s \) denoting that only the curvature part and only the flow straining part of flame stretch are involved, respectively. In general, the Karlovitz numbers in Eqs. (3.4) and (3.24) are not equal to each other: \( Ka_k \neq Ka'_c \neq Ka'_s \). However, the influence of each of these Karlovitz numbers is governed by a single Markstein number \( M = M'_c = M'_s \), which can be computed from the theoretical expression (3.19) or (3.20). The reason for this is the fact that the stretch rates \( K_k, K'_c, \) and \( K'_s \) are uniform in the flame for weak stretch, i.e. these stretch rates do not depend on the position within the flame to the lowest order, which indicates that the integrals in Eqs. (3.8) and (3.9) are similar for \( K_k, K'_c \) and \( K'_s \). The observation that the same Markstein number can be used for these different stretch rates has been mentioned by a number of authors before. For instance, Clavin and Williams [25], and Matalon and Matkowski [70] have derived this Markstein number for a general weakly distorted infinitely thin flame in which \( K'_c \) and \( K'_s \) are both included.

However, in literature (e.g. [17, 27, 47, 63]), the flame stretch rate is commonly separated in a different way as indicated in Eq. (3.23):

\[
K = K_c + K_s = \Sigma_{i=1}^{2} s_i^L / r_i + \Sigma_{i=1}^{2} \left( a_i + \frac{v_i^n}{r_i} \right),
\]

in which it is used that the flame propagation velocity \( v_i^f \) is the sum of the burning velocity \( s_i^L \) and the gas velocity normal to the flame surface \( v_i^n \) (cf. Eq. (1.1)). The total flame stretch rate in Eq. (3.25) is separated into a curvature part \( K_c = \Sigma_{i=1}^{2} s_i^L / r_i \) and a strain part \( K_s = \Sigma_{i=1}^{2} \left( a_i + \frac{v_i^n}{r_i} \right) \). The difference with the separation in Eq. (3.23) is that the straining of the flow relative to the curved flame surface \( v_i^n / r_i \) is considered as a straining effect rather than a curvature effect.

For the separation of the flame stretch rate as indicated in Eq. (3.25), Clavin and Joulin [27] proposed a similar linear combination of the curvature and flow straining part as in Eq. (3.24):

\[
\frac{m}{m_0} = 1 - Ka_c M_c - Ka_s M_s + \text{hot}.
\]

However, in general, the effects of curvature and strain can not be described by the same Markstein number \( M \neq M_c \neq M_s \) as is the case for the different
Markstein numbers in Eq. (3.24). In this section, it will be shown that these Markstein numbers are different and that the Markstein number for strain is not even unique. The reason is that the total flame stretch rate \( K \) is uniform throughout the flame, whereas the flame stretch rate for curvature \( K_c \) and strain \( K_s \) vary significantly throughout the flame.

In the remainder of this section, the separate Markstein numbers for curvature and strain as defined in Eq. (3.26) are studied. The special case of perfectly spherical flames is considered, in which the strain rates are equal to zero \( (\alpha_i = 0) \). The total flame stretch rate can then be written as:

\[
K(r) = K_c(r) + K_s(r) = \frac{2s_L(r)}{r} + \frac{2v'(r)}{r}. \tag{3.27}
\]

In order to be able to compute the Karlovitz integrals in Eqs. (3.8) and (3.9) for the separate contributions to the flame stretch rate \( K_c(r) = 2s_L(r)/r \) and \( K_s(r) = 2v'(r)/r \), it has to be known how these contributions depend on the radius of the sphere. As only lowest-order stretch is considered, the variables \( s(r) \), \( \rho^0(r) \), \( K(r) \), \( \phi^0(r) \), and \( T^0(r) \) in Eqs. (3.8) and (3.9) can be evaluated from stretchless information of a flat flame, as discussed in Section 3.3. Therefore, the burning and gas velocities in the curvature and straining parts of the stretch rate are found from flat stretchless flames. Also the radius at the inner layer is large compared to the flame thickness: \( r = r_{il}(1 + O(\delta^{0}_f/r_{il})) \). For weakly stretched spherical flames, the flame stretch rate in Eq. (3.27) reduces to:

\[
K = K_c + K_s = \frac{2s_L^0}{r_{il}} + \frac{2v_0^0}{r_{il}}. \tag{3.28}
\]

In Fig. 3.6, the behaviour of the burning velocity \( s_L^0(s) \), gas velocity \( v_0^0(s) \), and flame propagation velocity \( v_f^0(s) \) is schematically represented as a function of the position in different unstretched flat flames, with \( s \) the coordinate perpendicular to the flame surface. Fig. 3.6a shows the situation for a steady flame \( (v_f^0 = 0) \), Fig. 3.6b for a flame propagating into a closed tube \( (v_0^0(s_u) = 0) \), and Fig. 3.6c for a flame propagating out of a closed tube \( (v_0^0(s_b) = 0) \). As discussed above, for the weakly stretched spherical flames that are considered here, the velocity fields in Figs. 3.6a, b, and c can represent those of the steady, imploping, and expanding spherical flames, respectively. Notice that the flame propagation velocity is uniform throughout the flame in all cases since all flame parts move with the same velocity. However, the burning and gas velocities vary significantly throughout the flame due to density variations. Furthermore, the burning velocities show the same behaviour in all the different situations, which is not the case for the gas velocities.

### 3.4.2 Infinitely thin reaction layer

In this section, it is assumed that the reaction layer is infinitely thin. Therefore, the inner layer and the burnt flame boundary are considered to be located at the
3.4 Markstein numbers for curvature and strain

Figure 3.6: Burning, gas, and flame propagating velocities throughout unstretched flat flames which are steady (a), propagating into a tube (b), and propagating out of a tube (c), with \( s \) the coordinate perpendicular to the flame surface.

same position in the flame. Theoretical expressions for the Markstein numbers for both curvature and strain at the reaction layer are derived.

**Markstein number for curvature**

In order to find an expression for the Markstein number for the curvature part of the flame \( M_{b,c} \), \( \rho^0 K_c = 2m^0_b/r_b \) is substituted into the Karlovitz integrals in Eqs. (3.8) and (3.9), together with the expressions for \( \bar{T}^0_i(r) \) and \( \bar{\phi}^0_i(r) \) in Eqs. (B.12) and (B.13):

\[
IK^0_{b,c} = \frac{IK^0_{b,c}}{r_b} = \frac{2\bar{\delta}^0_i}{r_b} = \mathcal{K} a_{b,c}.
\]  

After inserting Eq. (3.29) into Eqs. (3.6) and (3.7) and using Eqs. (3.4) and (3.5), the Markstein number for the curvature part is obtained:

\[
M_{b,c} = 1 - \left( \frac{1}{Le_{fuel}} - 1 \right) \left( T^0_b - T_u \right) \frac{\partial \ln (m^0_b)}{\partial T^0_b}
\]

\[= 1 - \frac{2}{\varepsilon z_0} \left( \frac{1}{Le_{fuel}} - 1 \right), \tag{3.30}\]

where the temperature sensitivity of the mass burning rate that has been found by rate-ratio asymptotics is used (cf. Section 3.3.1). From Eq. (3.30), it can be concluded that the Markstein number for the curvature part \( M_{b,c} \) is well defined.
and not equal to the Markstein number for the total flame stretch $\mathcal{M}_b$ given by Eq. (3.19). The Markstein number for curvature is unique due to the fact that, although the stretch rate for the curvature $K_c$ varies rapidly in the flame, the combination of the stretch rate and density $\rho^0 K_c$ is uniform to the lowest order. The first expression in Eq. (3.30) has been found first by Barenblatt et al. [6] for the Markstein number of the total flame stretch, by neglecting the influence of density variations in the flame.

Markstein number for strain

The Markstein number for the flow straining part $\mathcal{M}_{b,s}$ follows from Eq. (3.26):

$$\mathcal{K}_{ab,s} \mathcal{M}_{b,s} = \mathcal{K}_{ab} M_b - \mathcal{K}_{ab,c} \mathcal{M}_{b,c}. \quad (3.31)$$

By using $v_0^b = s_{L,b}^0 + v_0^b$ and inserting the different contributions to the flame stretch rate in the Karlovitz numbers, these numbers become: $\mathcal{K}_{ab} = 2v_0^b \delta_s^0/s_{L,b}^0 r_b$, $\mathcal{K}_{ab,c} = 2\delta_s^0/r_b$, and $\mathcal{K}_{ab,s} = \mathcal{K}_{ab} - \mathcal{K}_{ab,c}$. Eq. (3.31) then reduces to:

$$v_0^b \mathcal{M}_{b,s} = v_0^b M_b + s_{L,b}^0 (M_b - \mathcal{M}_{b,c}). \quad (3.32)$$

It should be realised that the combined Markstein number $\mathcal{M}_b$ and the Markstein number for curvature $\mathcal{M}_{b,c}$ are well defined and given by Eqs. (3.19) and (3.30), respectively. Also the burning velocity at the burnt flame boundary $s_{L,b}^0$ has a constant value, which is independent of the combustion situation. Notice, however, that this is not the case for the gas velocity at this boundary $v_0^b$ (cf. Fig. 3.6). In Fig. 3.7a, the left-hand side of Eq. (3.32) $v_0^b \mathcal{M}_{b,s}$ is plotted as a function of the gas velocity scaled with the burning velocity at the burnt flame boundary $v_0^b/s_{L,b}^0$, thereby using the constant values $\mathcal{M}_b = 2.3$, $\mathcal{M}_{b,c} = 1.0$, and $s_{L,b}^0 = 2.5 \text{m/s}$ as an example.

An expression for the Markstein number for strain follows from Eq. (3.32):

$$\mathcal{M}_{b,s} = M_b + (\mathcal{M}_b - \mathcal{M}_{b,c}) \frac{s_{L,b}^0}{v_0^b}. \quad (3.33)$$

In Fig. 3.7b, the solid line represents the Markstein number for strain at different values for the gas velocity scaled with the burning velocity. Notice the variation of this Markstein number due to the different gas velocities at the burnt flame boundary. These different values indicate different combustion situations. The values $v_0^b/s_{L,b}^0 = -1, -\tau/(1 + \tau), 0$ in Fig. 3.7 represent a steady (cf. Fig. 3.6a), an imploding (cf. Fig. 3.6b), and an expanding (cf. Fig. 3.6c) spherical flame, respectively. For a steady flame, the Markstein numbers for strain and curvature are equal ($\mathcal{M}_{b,s} = \mathcal{M}_{b,c}$), which was already found by Clavin and Joulin [27] and Bradley et al. [17]. However, for imploding flames, the Markstein number for strain is smaller than the one for curvature ($\mathcal{M}_{b,s} < \mathcal{M}_{b,c}$). The Markstein
number for strain becomes even infinite for expanding flames, because the burnt gas velocity is zero. This is related to the observation that the expanding spherical flame is influenced by strain, although the burnt gas velocity is zero.

Fig. 3.7 indicates that the Markstein number for strain is not uniquely defined, because different combustion situations give different values for this Markstein number. Only the total Markstein number $M_b$ and the Markstein number for curvature $M_{b,c}$ are well defined and independent of the combustion situation. The Markstein number for strain $M_{b,s}$ depends on the flow throughout the flame. This contradicts with some previous studies, which make use of the assumption that different combustion situations can be used and combined to derive the Markstein numbers for strain and curvature separately. In [17, 47], this assumption is used to derive the separate Markstein numbers from experimental results of spherically expanding flames. However, the separation of the Markstein numbers in Eq. (3.26) as proposed by Clavin and Joulin [27] can only be used for studying and comparing results for each combustion situation separately.

### 3.4.3 Finite thickness of the reaction layer

In this section, theoretical expressions for the Markstein number for curvature and strain at the inner layer are derived for more realistic flames. In these flames, the reaction layer is considered to have a finite thickness. Therefore, the inner layer and the burnt flame boundary do not have the same position in the flame, as is the case in the previous section.

**Markstein number for curvature**

An expression for the Markstein number for curvature $M_{b,c}$ can be found by combining Eqs. (3.4), (3.5), and (2.12) and assuming the flame surface area to be
equal to one. The derivation is shown in Appendix B, in which the total flame stretch rate \( K \) in Eqs. (3.4), (3.5), and (2.12) has to be replaced by the flame stretch rate for curvature \( K_c = 2\sigma^0_L/r il \) (cf. Eq. (3.28)). The Markstein number for curvature at the inner layer then becomes:

\[
M_{il,c} = \frac{1}{\delta^0_T \rho^0 s^0_L} \left( \int_{s u}^{s b} \rho^0 s^0_L T^0 \, ds - \int_{s il}^{s b} \rho^0 s^0_L \, ds \right) - \left( T^0_b - T_u \right) \frac{\partial \ln \left( \frac{m^0}{T^0_b} \right)}{\partial T^0_b} \int_{s u}^{s b} \rho^0 s^0_L \left( \tilde{\phi}^0_{\text{fuel}} - T^0 \right) \, ds.
\]  

(3.34)

The unstretched density multiplied with the unstretched burning velocity \( \rho^0 s^0_L = m^0 \) is uniform throughout the flame. Therefore, Eq. (3.34) reduces to:

\[
M_{il,c} = \frac{1}{\delta^0_T} \left( \int_{s u}^{s b} \tilde{T}^0 \, ds - (s_b - s_{il}) \right) - \left( T^0_b - T_u \right) \frac{\partial \ln \left( \frac{m^0}{T^0_b} \right)}{\partial T^0_b} \int_{s u}^{s b} \rho^0 s^0_L \left( \tilde{\phi}^0_{\text{fuel}} - T^0 \right) \, ds.
\]  

(3.35)

Eq. (3.35) is only dependent on unstretched flame data \( \tilde{T}^0, T^0_b, \tilde{\phi}^0_{\text{fuel}}, \) and \( \delta^0 \) that are independent of the combustion situation. Therefore, the Markstein number for curvature is uniquely defined.

**Markstein number for strain**

In order to find an expression for the Markstein number for strain \( M_{il,s} \), the same procedure can be followed as in the derivation of Eq. (3.34). The only exception is that the flame stretch rate \( K \) has to be replaced by the flame stretch rate for strain \( K_s = 2v^0 / r il \) (cf. Eq. (3.28)) instead of the flame stretch rate for curvature:

\[
M_{il,s} = \frac{1}{\delta^0_T \rho^0 v^0 s^0 L} \left( \int_{s u}^{s b} \rho^0 v^0 \tilde{\phi}^0 \, ds - \int_{s il}^{s b} \rho^0 v^0 \, ds \right) - \left( T^0_b - T_u \right) \frac{\partial \ln \left( \frac{m^0}{T^0_b} \right)}{\partial T^0_b} \int_{s u}^{s b} \rho^0 v^0 \left( \tilde{\phi}^0_{\text{fuel}} - T^0 \right) \, ds.
\]  

(3.36)

The unstretched density multiplied with the unstretched gas velocity \( \rho^0 v^0 \) changes significantly throughout the flame. Furthermore, the unstretched gas velocity \( v^0 \) is different in different combustion situations (cf. Fig. 3.6). This indicates that
different Markstein numbers for strain can be found from Eq. (3.36) for different combustion situations. Therefore, the Markstein number for strain is not uniquely defined.

In this section, the special case of spherical flames has been discussed. Notice, however, that the argumentation about the non-uniqueness of the Markstein number for strain also holds for more generally curved and strained flames. The reason is that the strain rates can be taken into account in the Karlovitz number for strain that is used to derive Eqs. (3.31) and (3.36) without changing the rest of the argumentation significantly.

### 3.4.4 Theoretical and numerical results

In this section, the Markstein numbers for curvature and strain are computed from the theoretical expressions (3.30), (3.33), (3.35), and (3.36) for planar stagnation and perfectly spherical flames with equivalence ratios of $\phi = 0.8$ and $\phi = 1.0$. The unstretched data required to evaluate these theoretical expressions are the same as those used in Section 3.3.3. First, unit Lewis numbers are considered and, subsequently, non-unit Lewis numbers.

#### Unit Lewis numbers

It is assumed here that the Lewis numbers are equal to unity, which indicates that there is no preferential diffusion and that the Karlovitz integrals $IK^0_i$ and $IK^0_T$ are equal to each other. Therefore, Eqs. (3.35) and (3.36) for the Markstein numbers for curvature and strain reduce to:

\[
M_{il,c} = \int_{s_u}^{s_b} \left( \int_{s_b}^{T_0} ds - (s_b - s_{il}) \right) \tag{3.37}
\]

and

\[
M_{il,s} = \int_{s_u}^{s_b} \left( \int_{s_b}^{\rho \theta \nu \nu^0 ds - \int_{s_{il}}^{s_b} \rho \theta \nu \nu^0 ds \right), \tag{3.38}
\]

respectively.

In Table 3.4, the combined Markstein numbers and the Markstein numbers for curvature and strain are shown for $\phi = 0.8$ and $\phi = 1.0$. These Markstein numbers are computed from Eqs. (3.19), (3.30), and (3.33) in which the reaction layer is assumed to be infinitely thin and from Eqs. (3.22), (3.37), and (3.38) in which the finite thickness of the reaction layer is taken into account. Table 3.4 shows that the Markstein numbers derived for infinitely thin reaction layers are higher than the Markstein numbers derived for a finite thickness of the reaction layer. However, the information that can be deduced from these results is the same and confirms the findings in Sections 3.4.2 and 3.4.3. The combined Markstein number and the Markstein number for curvature are both unique. The Markstein number for strain is not unique and only the same for the steady spherical...
flames. The difference between these two flames is that one has the burnt gases in the centre of the flame ($v^f > 0$) and the other one has unburnt gases at that position ($v^f < 0$). For steady flames, the Markstein number for curvature and strain are equal. This was expected, because the mass burning rate is equal to the unstretched one in Eq. (3.26) and the Karlovitz numbers for curvature and strain are also equal but have opposite signs. Therefore, the corresponding Markstein numbers have to be equal as well. The combined Markstein number can not be determined for these flames, because the total stretch rate is zero. The stagnation flames endure no curvature and, therefore, the combined Markstein number and the one for strain are equal. For imploding flames, the results in Table 3.4 confirm the conclusion in Section 3.4.2 that the Markstein number for curvature is higher than the Markstein number for strain. Furthermore, the Markstein numbers for the stoichiometric flames are slightly higher than for the lean flames.

The Markstein number for flow straining that is obtained from Eq. (3.33) is infinite for spherically expanding flames, because the burnt gas velocity is zero. However, the Markstein number that is obtained from Eq. (3.38) has a finite value. This is due to the fact that the gas velocity is not evaluated at the burnt flame boundary anymore but at the inner layer where the gases are moving (cf. Fig. 3.6).

### Non-unit Lewis numbers

Equations (3.19), (3.30), (3.33), (3.20), (3.35), and (3.36) are used to compute the combined Markstein numbers and the Markstein numbers for curvature and strain.
3.5 Conclusions

In this chapter, a detailed investigation was performed on the relation between the mass burning rate and the flame stretch rate for weakly stretched flames. A number of new contributions were added to this subject.

The main contribution is the demonstration that the separation of the Markstein number in a part that describes the curvature of the flame and another part that describes the flow straining is not useful, because it is impossible to define a unique Markstein number for the flow straining that can describe its influence in different combustion situations.
Another contribution is the derivation of a relation to compute the Markstein number in which the structure of the flame is taken into account. The effort to compute the Markstein number from this relation is small, because only the structure of an unstretched flat flame is required. It was shown that the prediction of the Markstein number by this new relation is more accurate than the prediction by the asymptotic relations that can be found in literature.

Another finding is that the Markstein number appears to be strongly dependent on the position in the flame at which it is evaluated, because the burning velocity and the flame stretch rate are dependent on the position in the flame. The Markstein number can even have a different sign at the unburnt side of the flame than at the burnt side. In this chapter, the inner layer was chosen as the position in the flame to evaluate the Markstein number.

For small flame stretch rates, it is well known that the Markstein number is unique and can therefore predict the sensitivity of a flame to flame stretch for different kinds of flames such as flat stagnation flames and spherically expanding flames. However, for higher stretch rates, the Markstein number is unable to predict this sensitivity, because the relation between the mass burning rate and the flame stretch rate is not linear anymore. It was shown that, at these higher stretch rates, the flat stagnation flames deviate more from this linear behaviour than the spherically expanding flames at the same flame stretch rates.
4

Direct numerical simulations of freely expanding turbulent flames

4.1 Introduction

A DNS program is developed for the simulation of freely expanding flames in a turbulent flow field. In this chapter, details about this DNS program are given (cf. Sections 4.2, 4.3, 4.4, and 4.5), a validation is performed by simulating laminar flames (cf. Section 4.6), and simulations of turbulent flames are shown (cf. Section 4.7).

This DNS program is suitable to simulate different kinds of premixed expanding flames. However, this chapter focuses on stoichiometric atmospheric flames without preferential diffusion effects, which means that this study is restricted to unit Lewis numbers. These preferential diffusion effects are not taken into account, because stretch effects are of more interest here. Furthermore, the assumption of unit Lewis numbers makes the application of the used chemical reduction schemes less complex.

The computational domain has the shape of a cube and the discretisation of the combustion equations is performed in a cartesian coordinate system (cf. Section 4.2). Furthermore, a uniform mesh is used. The mesh size can affect the accuracy of the simulations, which is discussed in Section 4.5.

As in the previous chapters, flames in an open environment are the main subject of interest. This means that all boundaries of the cubic computational domain have to be modelled as outlet boundaries to prevent pressure build-up in the domain. Most already existing DNS programs have been developed to simulate flat flames in a turbulent environment (cf. e.g. [7, 37, 88, 89, 97, 110]). These programs not only contain an outlet boundary, but also inlet and lateral boundary conditions. An exception is the DNS program of Jenkins and Cant [52], which also has been developed to simulate turbulent expanding flames in an open environment. Modelling of all boundaries as outlet boundaries is not common and gives rise to additional problems. Therefore, extra attention is paid to these boundary conditions in Section 4.3.
The computational costs of DNS are extremely high. These costs are reduced here by using a small computational domain. Furthermore, two distinct chemical reduction techniques are used to describe the chemistry instead of the use of a detailed reaction mechanism: the single-step reaction mechanism and the Flamelet-Generated Manifold (FGM) method (cf. Section 4.4). The single-step reaction mechanism is a commonly used mechanism in DNS programs. However, this mechanism requires several coefficients to be fitted to a specific combustion situation, which makes this mechanism less universally applicable. To overcome this disadvantage, a different reduction technique is also applied here, which is the recently developed FGM method (cf. [79]).

The DNS program is validated by simulating flat stretchless flames and laminar spherically expanding flames and comparing the results to flame simulations with a one-dimensional reference program (cf. Section 4.6). Due to the very small computational domain, the laminar spherically expanding flames have very small flame radii and are therefore strongly stretched, which makes these test cases very challenging. Finally, various turbulent flow fields are used to investigate the flame response to these turbulent fields (cf. Section 4.7).

4.2 Discretisation

A compact finite difference method is used for the discretisation of the combustion equations in space which are the continuity equation (1.16), Navier-Stokes equations (1.43), temperature equation (1.45), and mass balance equations (1.46). These equations require approximations of the first and second order derivatives, which are given in Section 4.2.1. The time integration is performed with a third-order Runge-Kutta method. This method is explained in Section 4.2.2. The computations are performed on a collocated mesh instead of a staggered mesh. Furthermore, parallel computing and data transfer is implemented through the Message Passing Interface (MPI).

4.2.1 Spatial discretisation

The one-parameter family of tridiagonal schemes is used for the spatial discretisation [62]. For a one-dimensional uniformly spaced mesh, the finite difference approximations \( f'_k \) and \( f''_k \) of the first and second derivatives \( df/dx(x_k) \) and \( d^2f/dx^2(x_k) \) of a general function \( f(x) \) at mesh point \( k \) are given by:

\[
\alpha_1 f'_{k-1} + f'_k + \alpha_1 f'_{k+1} = a_1 \frac{f_{k+1} - f_{k-1}}{2\Delta x} + a_2 \frac{f_{k+2} - f_{k-2}}{4\Delta x},
\]

and

\[
\alpha_2 f''_{k-1} + f''_k + \alpha_2 f''_{k+1} = a_3 \frac{f_{k+1} - 2f_k + f_{k-1}}{(\Delta x)^2} + a_4 \frac{f_{k+2} - 2f_k + f_{k-2}}{4(\Delta x)^2}.
\]
respectively, with
\[ a_1 = \frac{2}{3} (\alpha_1 + 2), \quad a_2 = \frac{1}{3} (4\alpha_1 - 1), \]
\[ a_3 = \frac{4}{3} (1 - \alpha_2), \quad a_4 = \frac{1}{3} (-1 + 10\alpha_2), \]
and \( \Delta x \) the mesh size. A linear system of equations for the unknown derivative values is formed by writing the relations (4.1) and (4.2) for each mesh point. This system is cyclic tridiagonal if the boundaries are periodic. Otherwise, additional relations for the mesh points on and near the boundaries are required (cf. Section 4.3).

From the scheme in (4.1) and (4.2), the fourth-order central difference scheme [50] is recovered in the case that \( \alpha_1 = 0 \) and \( \alpha_2 = 0 \) and the classical Padé scheme [28] is found in the case that \( \alpha_1 = 1/4 \) and \( \alpha_2 = 1/10 \). Both these schemes are fourth-order accurate. However, in the case that \( \alpha_1 = 1/3 \) and \( \alpha_2 = 2/11 \), the truncation error coefficient of the leading order disappears which causes the scheme in (4.1) and (4.2) to become formally sixth-order accurate. This scheme is called a compact finite difference scheme [62] and is employed here.

Compared to the traditional finite difference schemes, a compact finite difference scheme provides a better representation of the shorter length scales. Therefore, the compact scheme is especially suitable for solving problems with a broad range of spatial scales as in turbulent flows. This property brings the compact scheme close to the spectral methods without having some of the disadvantages of the spectral methods such as the restriction to simple mesh geometries and periodic boundary conditions.

Due to the symmetrical definition of the derivatives in Eqs. (4.1) and (4.2), the non-linear convection terms can amplify the grid oscillations in the three-dimensional computations. To avoid this amplification, an explicit fifth-order upwind scheme is used to discretise the convection terms in the combustion equations, cf. [8]. A fifth-order scheme is chosen in order to maintain a high accuracy on the one hand and keep the required stencil relatively small on the other hand. In the event that the flow is directed in the positive increasing \( k \)-direction, the applied upwind scheme is written as:
\[ f'_k = \frac{1}{\Delta x} \left( -\frac{1}{30} f_{k-3} + \frac{1}{4} f_{k-2} - f_k - \frac{1}{3} f_{k+1} + \frac{1}{2} f_{k+3} - \frac{1}{20} f_{k+2} \right), \quad (4.4) \]
for a one-dimensional uniformly spaced mesh.

### 4.2.2 Temporal discretisation

The time integration is performed with a compact storage third-order Runge-Kutta method derived by Wray [116]. To take a step in time \( \Delta t \), this method requires three evaluations of the right-hand side of the general expression
\[
\frac{dx}{dt} = f(x, t), \quad (4.5)
\]
which can represent the combustion equations in Section 1.3. The time integration applied to Eq. (4.5) is divided into the following substeps:

\[
\begin{align*}
    x' &= x^m + a_1 f(x^m, t^m) \Delta t, \\
    x'' &= x^m + a_2 f(x^m, t^m) \Delta t, \\
    x''' &= x' + a_3 f(x'', t'') \Delta t, \\
    x^{m+1} &= x' + a_4 f(x''', t''') \Delta t,
\end{align*}
\]

(4.6)

with superscript \( m \) the time index,

\[
    a_1 = \frac{1}{4}, \quad a_2 = \frac{8}{15}, \quad a_3 = \frac{5}{12}, \quad a_4 = \frac{3}{4},
\]

(4.7)

and intermediate times

\[
    t' = t^m + \frac{1}{4} \Delta t, \quad t'' = t^m + \frac{8}{15} \Delta t, \quad t''' = t^m + \frac{2}{3} \Delta t.
\]

(4.8)

The stability of the time advancement depends on both the time integration scheme and the spatial discretisation [116]. Notice that the time step is limited by the travelling velocity of acoustic waves \( c \):

\[
    \Delta t \leq \frac{\Delta x}{c}.
\]

(4.9)

\section*{4.3 Boundary conditions}

The DNS program uses algorithms that have very low numerical dissipation. Therefore, it requires precise boundary conditions to avoid numerical instabilities and to control spurious wave reflections at the computational boundaries. The choice was made to use the formulation of the boundary conditions developed by Poinset and Lele [91, 92] instead of the more classical methods such as the use of extrapolation and Riemann invariants (cf. e.g. [96]), because the formulation of Poinset and Lele has been proven to perform better in most cases. Their method is referred to as Navier-Stokes Characteristic Boundary Conditions (NSCBC) and it was developed for non-reactive flows (cf. [92]) and reactive flows with simple chemistry (cf. [91]). Baum \textit{et al.} [9] adapted the NSCBC method to be able to describe reactive flows that are modelled with a detailed reaction mechanism and realistic thermodynamic properties. However, this extension of the NSCBC method is not implemented in the DNS program, because the computations of the expanding flames are stopped before the flame reaches the computational boundaries. The NSCBC method can be used to model reflecting and non-reflecting boundaries, inlet and outlet boundaries, slip and no-slip walls, and subsonic and supersonic flows.
The NSCBC method is based on the inviscid characteristic theory for the Euler equations [109], which are hyperbolic equations. It is known that every system of hyperbolic equations can be associated with a corresponding system of propagating waves. At the boundaries of a computational domain two kinds of waves exist: the ones leaving the domain and the ones entering the domain. The waves going out of the domain are completely determined by the solution inside the domain. The waves coming into the domain are not determined and therefore have to be specified by a proper set of boundary conditions. The NSCBC method uses the outgoing waves to deduce these boundary conditions. However, the Navier-Stokes equations are not hyperbolic due to the viscous terms. The NSCBC method treats these extra terms by supplying additional conditions.

A concise description of the NSCBC method is given below. A more comprehensive description can be found in [90–92]. Using the characteristic analysis, the governing equations can be decomposed into an equivalent system in which the characteristic waves can be identified. This system is symmetrical for the different directions \(x, y,\) and \(z\). If the boundary conditions associated with the \(x\) direction are considered, the continuity equation, the Navier-Stokes equations, the temperature equation, and the mass-balance equation for each species can be reformulated as:

\[
\frac{\partial \rho}{\partial t} + \frac{1}{c^2} \left( L_2 + \frac{1}{2} (L_5 + L_1) \right) + C_1 = 0, \tag{4.10}
\]

\[
\frac{\partial u}{\partial t} + \frac{1}{2\rho c} (L_5 - L_1) + C_2 = 0, \tag{4.11}
\]

\[
\frac{\partial v}{\partial t} + L_3 + C_3 = 0, \tag{4.12}
\]

\[
\frac{\partial w}{\partial t} + L_4 + C_4 = 0, \tag{4.13}
\]

\[
\frac{\partial T}{\partial t} + \frac{T}{\rho c^2} \left( -L_2 + \frac{1}{2} (\gamma - 1) (L_5 + L_1) \right) + C_5 = 0, \tag{4.14}
\]

\[
\frac{\partial Y_i}{\partial t} + L_5 + L_{5+i} + C_{5+i} = 0, \tag{4.15}
\]

in which \(u, v,\) and \(w\) are the velocity components in the cartesian coordinate system, \(\gamma\) is the specific heat ratio, and the \(C\)'s are the terms that do not contain the first derivative of a primitive variable along the \(x\) direction. These terms are the transverse, viscous, diffusive, and reactive terms. The \(L\)'s in the above equations represent the amplitudes of the characteristic waves crossing the boundaries:

\[
L_1 = \lambda_1 \left( \frac{\partial \rho}{\partial x} - \rho c \frac{\partial u}{\partial x} \right), \tag{4.16}
\]

\[
L_2 = \lambda_2 \left( c^2 \frac{\partial \rho}{\partial x} - \frac{\partial p}{\partial x} \right), \tag{4.17}
\]

\[
L_3 = \lambda_3 \frac{\partial v}{\partial x}, \tag{4.18}
\]
where the λ's are the characteristic velocities: \( \lambda_1 = u - c \), \( \lambda_2 = \lambda_3 = \lambda_4 = \lambda_{5+i} = u \), and \( \lambda_5 = u + c \). By neglecting the C's in Eqs. (4.10)–(4.15), a local one-dimensional inviscid (LODI) system of equations is obtained. These LODI relations contain information about the wave structure and the propagation direction of the characteristic waves. Therefore, these relations can be used to obtain expressions for the amplitude of the incoming characteristic waves in terms of the outgoing waves which can be determined by information inside the domain. These waves from the inviscid one-dimensional relations are assumed to have the same amplitude as the waves from the viscous Navier-Stokes problem.

For each physical condition that fixes a primitive variable on the boundary, the corresponding equation in the set of governing equations (4.10)–(4.15) does not have to be solved at the boundary. In the remaining equations, the expressions for the incoming waves are inserted and also additional conditions are specified to handle the viscous and diffuse terms in the equations.

For example, if an adiabatic no-slip wall is present, the physical conditions are that the heat flux and the three velocity components at the boundary are zero. Therefore, Eqs. (4.11), (4.12), and (4.13) do not have to be solved at the boundary. The characteristic waves in the remaining equations (4.10), (4.14), and (4.15) can be found from the LODI relations. These relations show that \( L_3 = L_4 = 0 \) and that the incoming wave \( L_1 \) can be expressed in terms of the outgoing wave \( L_5 \): \( L_1 = L_5 \), in which \( L_5 \) can be computed from the values of the variables at the grid points inside the domain. Furthermore, the characteristic waves \( L_2 \) and \( L_{5+i} \) are also zero, because the velocity normal to the boundary is zero. The remaining equations (4.10), (4.14), and (4.15) can be solved at the no-slip adiabatic wall by inserting the above expressions for the characteristic waves and by setting the normal heat flux and the normal reactant diffusion flux to zero. Several other applications of these boundary conditions can be found in [7].

The DNS program is developed to simulate expanding flames in an open environment. Therefore, non-reflecting outlet boundaries are required, which means that the gases are able to leave the domain and that the acoustic waves are not reflected at the computational boundaries. Furthermore, if turbulent flows are considered, the turbulent vortices have to convect out of the domain without leaving their influence at the interior of the domain. Notice that no boundary conditions would be required, if periodicity at the computational boundaries is assumed. In that case, the computational domain is folded onto itself. This also means that the pressure in the computational domain increases due to the exothermic reactions that take place inside the domain. However, the effect of these pressure variations on the propagation of these flames is not of interest here.
To obtain a perfectly non-reflecting outflow, a single physical boundary condition should be specified, because only one characteristic wave $L_1$ is entering the domain. The other characteristic waves can be computed from information inside the domain. At a non-reflecting boundary, the incoming wave is modelled by $L_1 = 0$. However, prescribing all boundaries as perfectly non-reflecting outflow boundaries can lead to an ill-posed problem, because the mean pressure of the flow is not determined. The solution proposed by the NSCBC method is to use partially non-reflecting boundary conditions by implicitly imposing the pressure at infinity by the specification of incoming acoustical waves. In this way, information about the mean pressure is allowed to travel from regions far from the computational domain to the inside of this domain. To impose this condition, the incoming wave $L_1$ is modelled by:

$$L_1 = K_p (p - p_{at}),$$

(4.22)

where $p_{at}$ is the atmospheric pressure at infinity and $K_p$ is the pressure relaxation constant, which can be computed from $K_p = C_p (1 - Ma^2) c/L$. The constant $C_p$ is chosen to be 0.58 (cf. [92]), $L$ is the characteristic length of the domain, and $Ma$ is the Mach number. This condition for the pressure does not fix a variable at the boundary and, therefore, all of the combustion equations have to be solved at the boundary. Furthermore, the viscous boundary conditions require the spatial derivatives of the tangential stresses, the normal heat flux, and the normal reactant diffusion flux to be zero in the combustion equations at the boundary. Notice that the DNS program uses centered differences with a five-point numerical stencil. These central differences are replaced by a one-sided discretisation scheme with a three-point numerical stencil on and near the boundaries, because information is only available inside the domain.

### 4.4 Chemical reduction methods

The use of a detailed reaction mechanism to describe the chemistry involves the computation of a large and stiff set of mass-balance equations, which induces very high computational costs. In order to make the computations with the DNS program affordable, two different chemical reduction techniques are used: the single-step reaction mechanism and the Flamelet-Generated Manifold (FGM) method. These techniques are discussed in Sections 4.4.1 and 4.4.2, respectively. As an evaluation of both these reduction methods, stretched flames were simulated with these methods and compared to simulations using a detailed reaction mechanism (cf. Section 4.4.3). These simulations were performed with the flamelet method that is described in Chapter 2.

#### 4.4.1 Single-step reaction mechanism

The single-step reaction mechanism is based on the modelling of the global reaction only instead of the extensive whole set of elementary reactions that occur in
the combustion process. The omission of the intermediate species can be justified by the fact that these species are very rapidly formed and consumed in a thin reaction layer. The main advantage of this mechanism is the large reduction of the computational costs in comparison with more detailed reaction mechanisms, because less mass-balance equations (cf. Eq. (1.46)) have to be solved. The main disadvantage is that the coefficients in the expression for the mass consumption rate have to be fitted to a specific flame situation, which makes the method less universally applicable (cf. [65]).

In this study, methane-air flames are considered. The global reaction of these flames is presented in Eq. (1.37). The mass consumption rates of the four species involved (methane CH\(_4\), oxygen O\(_2\), carbon dioxide CO\(_2\), and water H\(_2\)O) are related to each other by this global reaction, for example \(\rho \cdot \text{O}_2 = 2M_{\text{O}_2}/M_{\text{CH}_4}\). Therefore, an expression for the mass consumption rate of methane is sufficient to solve the four mass-balance equations corresponding to the four species. In the case of flames with unit Lewis numbers, the mass fractions of the four species are directly related to each other, which means that only one mass-balance equation has to be solved. Notice that methane-air flames also contain nitrogen N\(_2\). However, this species is considered not to react with the other species. To ensure the conservation of mass, the mass fraction of nitrogen \(Y_{\text{N}_2}\) is computed from \(\sum_{i=1}^{N_i} Y_i = 1.0\).

The mass consumption rate of methane \(\dot{\rho}_{\text{CH}_4}\) can be written as an Arrhenius-like expression (cf. Section 1.3.3):

\[
\dot{\rho}_{\text{CH}_4} = -A_c \rho^{\alpha_{\text{CH}_4}} \chi^{\alpha_{\text{CH}_4}} \chi^{\alpha_{\text{O}_2}} Y_{\text{O}_2} e^{-E_a/RT},
\]

with \(\alpha_{\text{CH}_4}\) and \(\alpha_{\text{O}_2}\) the reaction orders of methane and oxygen, respectively. Next, it is briefly explained how the coefficients \(A_c\), \(\alpha_{\text{CH}_4}\), \(\alpha_{\text{O}_2}\), and \(E_a\) in Eq. (4.23) can be obtained. The resulting values for these coefficients are given for the case that is considered in this chapter, which is a stoichiometric flame with unit Lewis numbers. The coefficient \(A_c\) was obtained from fitting the mass burning rate to the unstretched mass burning rate \(m_0^i\) from adiabatic one-dimensional flame simulations that were performed with GRI-mech 3.0 [14], which is a detailed reaction mechanism that contains 325 reactions and 53 species. This unstretched mass burning rate \(m_0^i\) and the corresponding coefficient \(A_c\) have values of 0.3161 kg/m\(^2\)/s and 0.14269 \(\times\) \(10^{11}\) m\(^3\)/kg s, respectively. The reaction was chosen to be second-order, with \(\alpha_{\text{CH}_4} = 1.0\) and \(\alpha_{\text{O}_2} = 1.0\), in order to obtain a flame thickness that corresponds to flame simulations performed with GRI-mech 3.0. The activation energy was obtained by fitting the flames to heat losses: \(E_a = 1.372 \times 10^5\) J/mol (cf. [60,65]).

The backward reaction of the global reaction in Eq. (1.37),

\[
\text{CO}_2 + 2\text{H}_2\text{O} \rightarrow \text{CH}_4 + 2\text{O}_2,
\]

is not modelled, because its reaction rate is very small in comparison with the reaction rate of the forward reaction. Therefore, the backward reaction has no significant influence on the mass burning rate. Only the chemical equilibrium in
the burnt gases is influenced by the backward reaction, which causes the adiabatic temperature to become higher if the backward reaction is not modelled.

Also, the adiabatic temperature predicted with the single-step reaction mechanism is higher than the one predicted with more detailed reaction mechanisms. The main reason is that the conversion of carbon monoxide into carbon dioxide is not modelled with the single-step mechanism, which causes the chemical equilibrium to be different. It is possible to correct for this difference by using a patch factor that acts on the heat release (cf. e.g. [10]) or to incorporate carbon monoxide into the global reaction (cf. e.g. [36]). However, the choice was made not to correct for this difference, because it is not the goal here to predict the adiabatic flame temperature with a high level of accuracy.

4.4.2 Flamelet-generated manifold method

The Flamelet-Generated Manifold (FGM) method is a new reduction technique that was developed by van Oijen [79]. The basic idea of this method is to use one-dimensional flame simulations to compute multi-dimensional flames. The FGM method is based on two well-known reduction techniques that have been proven to be well suited for different kinds of flame simulations: the flamelet approaches (cf. e.g. Chapter 2) and the Intrinsic Low-Dimensional Manifold (ILDM) method (cf. e.g. [12, 66, 67]). As in the flamelet approaches, the FGM method assumes that a multi-dimensional flame can be constructed from a set of one-dimensional flames, because the inner structure of these flames are assumed to be similar. However, the implementation of the FGM method in the flame simulations is similar to the ILDM method. The FGM method has been proven to very accurately reproduce simulations with a detailed reaction mechanism of laminar premixed flames and partially premixed flames such as triple flames (cf. [79]). These FGM simulations have been shown to be computationally far less expensive than the simulations with a detailed reaction mechanism. A short explanation of the FGM method is presented below. For more detailed information on the FGM method and its applications, the reader is referred to [76–78].

The FGM method consists of two parts. One part is the generation of a manifold from flamelet computations and the other part concerns the use of this manifold in flame simulations.

A manifold is stored in a database which contains several variables such as the reaction rates as a function of a controlling or progress variable. To obtain a unique mapping of the variables onto the progress variable, this progress variable \( \mathcal{Y} \) can be one of the species mass fractions or a linear combination of these mass fractions that obeys \( \nabla \mathcal{Y} \neq 0 \). The manifold is generated by solving the one-dimensional flamelet equations that are shown in Eqs. (2.12), (2.13), and (2.14) in which multi-dimensional perturbations are neglected, i.e. the flame stretch rate \( K \) is zero in these equations. This procedure generates a one-dimensional manifold, which means that the manifold forms a curve in the composition space from the unburnt mixture to the chemical equilibrium mixture, which relates all variables to one progress variable. It is also possible to generate a multi-dimensional mani-
ifold, which spans a larger part of the composition space. A multi-dimensional manifold can be constructed by solving the one-dimensional flamelet equations at various compositions of the unburnt mixture.

The other part of the FGM method is the use of the manifold to perform the flame simulations. The set of equations that has to be solved consists of the continuity equation (1.16), the Navier-Stokes equations (1.43), the enthalpy equation (1.44) or the temperature equation (1.45), and of the differential equation for the progress variable $Y$:

$$\frac{\partial (\rho Y)}{\partial t} + \nabla \cdot (\rho v Y) = \nabla \cdot \left( \frac{\lambda}{L e y c_p} \nabla Y \right) + \rho_y,$$

which has a structure identical to the mass-balance equations (1.46). Notice that the mass-balance equations do not have to be solved, because the mixture composition is determined by the progress variable. The variables in this set of equations such as the reaction rates and the specific heat at constant pressure are obtained from the one-dimensional manifold and are used to solve this set of equations. In case a multi-dimensional manifold is used, extra differential equations have to be solved, because each additional dimension introduces an additional progress variable and corresponding differential equation. Therefore, the use of a multi-dimensional manifold increases the computational time. On the other hand, it also increases the accuracy of the flame simulations.

The two main reasons for the large reduction of the computational time in comparison with computations with a detailed reaction mechanism are the large reduction of the number of differential equations that have to be solved, and the larger time steps that can be taken due to the elimination of the smallest time scales. It should be noticed that the construction of a manifold in itself can also be time consuming. However, a manifold can be used for several flame simulations. A new manifold has to be constructed only in the case of different circumstances, such as for flames that contain another equivalence ratio or the use of another chemical reaction mechanism.

For the adiabatic and stoichiometric flames with unit Lewis numbers that are considered in this study, it appeared that the use of a one-dimensional manifold instead of a more-dimensional one is sufficient to simulate these flames accurately, because the enthalpy and the element mass fractions do not vary. Therefore, one-dimensional manifolds were constructed here. The choice was made to use the mass fraction of carbon dioxide as the progress variable, because this mass fraction is continuously increasing going from the unburnt to the burnt side of the flame.

In this chapter, two kinds of FGM simulations are performed: simulations with a manifold based on the single-step reaction mechanism and based on the detailed reaction mechanism GRI-mech 3.0 [14]. In order to validate the FGM method in the DNS program, the FGM simulations with a manifold based on the single-step reaction mechanism are compared to the simulations with the single-step reaction mechanism (cf. Section 4.4.1).
4.4.3 Evaluation of the reduction methods

In this section, the response of stretch on flames computed with the two reduction techniques described in Sections 4.4.1 and 4.4.2 are compared with each other, and with the detailed reaction mechanism GRI-mech 3.0. For this purpose, spherically expanding flames were simulated with the flamelet method that is described in Chapter 2, because this flamelet model has been proven to give very accurate results. The one-dimensional manifolds of the FGM method are based on the single-step reaction mechanism and on GRI-mech 3.0. The results are shown in Fig. 4.1 for a stoichiometric flame with unit Lewis numbers. In this figure, the ratio of the unstretched and stretched mass burning rate is plotted as a function of the Karlovitz number (cf. Eq. (3.3)) at the inner layer.

Figure 4.1: The ratio of the unstretched and stretched mass burning rate as a function of the Karlovitz number at the inner layer obtained from flamelet simulations (cf. Chapter 2) of spherically expanding flames. Solid line: GRI-mech 3.0, dashed line: the FGM method based on GRI-mech 3.0, dotted line: the FGM method based on the single-step reaction mechanism, dash-dotted line: the single-step reaction mechanism. Note that the dotted and dash-dotted lines coincide.

Fig. 4.1 shows that the computations with the single-step reaction mechanism and with the FGM method predict almost the same influence of the flame stretch on the mass burning rate as the computations with GRI-mech 3.0 do. The mass burning rate is only slightly overestimated by the computations with the reduction techniques. The difference in the ratio of the unstretched and stretched mass burning rate at a Karlovitz number of 0.08 is less than 0.4%. The results are almost identical for the computations with the single-step reaction mechanism and with the FGM method based on this single-step mechanism. Therefore, the dotted line in Fig. 4.1 is hardly visible.

4.5 Mesh size

In order to represent the small length scales in the flame simulations correctly, such as the structure of the inner layer, the demands on the mesh size are very
restrictive. These demands can be even more restrictive than those for the representation of the Kolmogorov length scale, which is the smallest turbulence length scale, because the thickness of the inner layer can be smaller than this turbulence scale. The structure of the inner layer is dependent on the representation of the source terms of the temperature \( S_T := \sum_{i=1}^{N_s} h_i \rho_i \) and the mass fractions \( S_Y := \rho_i \).

For the single-step reaction mechanism with unit Lewis numbers, the normalized source terms of the temperature and mass fractions are identical: \( \tilde{S}_T = \tilde{S}_Y \).

In Fig. 4.2, the normalized source term of the temperature is plotted through a flat stretchless stoichiometric flame with unit Lewis numbers computed with the single-step reaction mechanism.

\[
\begin{align*}
\tilde{S}_T &:= \frac{S_T}{S_Y} \\
\tilde{S}_Y &:= \frac{S_Y}{S_Y} \\
\end{align*}
\]

Figure 4.2: The normalized source term of the temperature \( \tilde{S}_T \) as a function of the coordinate perpendicular to the flame surface computed with the single-step reaction mechanism and for \( \phi = 1.0 \) and \( Le_i = 1 \).

The representation of the source terms influences the mass burning rate, which is one of the main parameters of interest. This influence is shown in Fig. 4.3 for an adiabatic and stretchless flat premixed flame that moves through various one-dimensional computational domains with uniformly spaced meshes. This flame is stoichiometric with unit Lewis numbers and the single-step reaction mechanism presented in Section 4.4.1 is used. For these flame simulations, an instationary numerical solver was used. This solver contains a finite difference approach which is second-order accurate [42]. Fig. 4.3 shows the lower and upper limits of the relative error of the mass burning rate \( \varepsilon \) as a function of the mesh size \( \Delta x \) of the computational domain. This relative error is defined as \( \varepsilon := (m_{\Delta x} - m^0) / m^0 \) in which \( m_{\Delta x} \) is the mass burning rate computed on a grid with a mesh size of \( \Delta x \), and \( m^0 = 0.3161 \text{kg/m}^2\text{s} \) which is obtained from simulations with an adaptive grid and very small mesh sizes. For a flame simulation on a certain mesh, the mass burning rate and therefore the relative error varies between the lower and upper limits shown in Fig. 4.3. This variation is caused by the way in which the source terms are reproduced by the mesh points while the flame moves through the computational domain. This study on the mesh size was performed on one-dimensional computational domains. However, the results can also be applied to two-dimensional and three-dimensional domains. In that case the maximum
4.6 Validation of the DNS program

The validation of the DNS program is performed by simulating laminar flames and comparing the results to simulations with a program called CHEM1D. This program can perform direct numerical simulations of one-dimensional flames and it has been proven to give very accurate results. More information about CHEM1D can be found in [42]. The considered laminar flames are flat stretchless flames (cf. Section 4.6.1) and expanding flames that are perfectly spherical (cf. Section 4.6.2). These flames are stoichiometric with unit Lewis numbers.

4.6.1 Flat flames

As a first validation of the DNS program, flat stretchless adiabatic flame simulations are compared to flame simulations with the reference program CHEM1D. The flat flames are propagating in the $x$ direction of the computational domain.

Figure 4.3: The lower (dashed line) and upper (solid line) limits of the relative error of the mass burning rate $\epsilon = (m^{1\Delta x} - m^0)/m^0$ as a function of the mesh size $\Delta x$ obtained from simulations of flat flames that propagate through the computational domain and for $\phi = 1.0$ and $Le = 1$. These simulations were performed with the single-step reaction mechanism and with the numerical solver that is described in [42].
Therefore, the combustion variables do not vary in the \( y \) and \( z \) directions. The length of the computational domain in the \( x \) direction is 100 mm. Three distinct meshes are considered with 1001, 2001, and 4001 grid points, which means that the mesh sizes are 0.1, 0.05, and 0.025 mm, respectively. The other two directions each contain five grid points with an identical mesh size as in the \( x \) direction. These two directions are modelled with periodic boundary conditions, whereas the \( x \) direction has an inlet boundary of unburnt gases at the left side of the computational domain and an outlet boundary of burnt gases at the other side. This outlet boundary is partially non-reflecting and implemented as described in Section 4.3. At the inlet boundary, the gas velocity and the temperature are imposed. The unburnt burning velocity is chosen as the inlet gas velocity, which prevents the flame from moving through the computational domain. The flame is located at approximately 20 mm from the inlet and the combustion variables of the initial flame structure are obtained from the simulations with CHEM1D.

Besides the use of this DNS program in its full three-dimensional form, it was also adapted to a single-dimension version. As expected, both versions proved to give identical results.

In the simulations with the DNS program, the chemistry is modelled with the single-step reaction mechanism as described in Section 4.4.1 and with the FGM method as described in Section 4.4.2. The FGM method uses two different manifolds, which are based on the single-step reaction mechanism and on GRI-mech 3.0. These simulations were compared with simulations with CHEM1D in which the chemistry is modelled with the single-step reaction mechanism and GRI-mech 3.0.

For the DNS computations with the smallest mesh size \( \Delta x = 0.025 \) mm, the mass burning rate is predicted within 0.1% accuracy compared to the results from CHEM1D in which an adaptive grid is used. This very small disagreement is due to both the difference in the used mesh and the DNS program being fully compressible, whereas CHEM1D uses the low-Mach number approximation (cf. [20]) which means that the pressure is assumed to be uniform. This approximation is valid for the studied flames, because the pressure is less than 1 Pa higher at the unburnt side of the flame than at the burnt side (cf. Fig. 4.4). The difference in the mass burning rate is smaller for the computations with the FGM methods than with the single-step reaction mechanism. This is probably due to the fact that the manifolds of the FGM methods are obtained from CHEM1D computations.

Furthermore, as is shown in Section 4.5, a large mesh size can cause the mass burning rate to deviate from its actual value and can cause fluctuations of the mass burning rate as the flame moves through the computational domain. It appeared that smaller mesh sizes cause the mass burning rate of the DNS computations to converge faster and fluctuate less than the mass burning rate of the CHEM1D computations, because the DNS program uses more accurate discretisation schemes (cf. Section 4.2) than CHEM1D, whose schemes are second-order accurate.

Besides the agreement between the mass burning rates obtained from the different numerical programs and the different chemical models, other characteris-
tics of the flame such as its temperature and inner structure are in very good correspondence as well. The oxidation layers obtained from the simulations using the FGM method based on GRI-mech 3.0 and those coming from GRI-mech 3.0 itself have a similar structure and are larger than the oxidation layers obtained from using the single-step reaction mechanism and the FGM method based on this mechanism. The larger oxidation layer is due to the oxidation reactions in the detailed reaction mechanism GRI-mech 3.0, which are not present in the single-step reaction mechanism.

The initial flame conditions of the DNS computations are obtained from computations with CHEM1D. Therefore, the initial pressure is constant and atmospheric in the whole computational domain. However, the fully compressible DNS program computes a more realistic pressure field in which the pressure slightly reduces going from the unburnt to the burnt side of the flame. Fig. 4.4 shows the pressure in the whole domain at \( t = 0.25, 0.30, 0.35, \) and 1.00ms after the start of the simulation. The flame is present at approximately \( x = 20 \text{ mm} \). Initially, the flame induces pressure waves which travel from the flame into the
unburnt and burnt side of the domain. These pressure waves are acoustic waves travelling with the velocity of sound. Notice that the velocity of sound is higher for the burnt gases than for the unburnt gases \((c = \sqrt{\gamma RT})\). The boundary conditions are modelled such that these waves reflect at the inlet boundary \((x = 0\text{ mm})\) and leave the domain at the outlet boundary \((x = 100\text{ mm})\). The outlet boundary is modelled as being partially non-reflecting (cf. Section 4.3), which causes a very small reflection back into the domain. The large density variation in the flame causes the pressure waves coming from the inlet boundary to partially reflect at the flame back to the inlet. Fig. 4.4 also shows that the pressure waves from simulations with the single-step reaction mechanism and with the FGM method based on this mechanism behave similarly, in spite of the fact that the manifold of the FGM method is based on CHEM1D computations.

### 4.6.2 Spherically expanding flames

As a second test case for the DNS program, perfectly spherical expanding flames are simulated using the single-step reaction mechanism (cf. Section 4.4.1) and the FGM method (cf. Section 4.4.2) which uses two different manifolds based on the single-step reaction mechanism and on GRI-mech 3.0. These flames are computed in a cubic three-dimensional computational domain with a length of 5.0 mm and 127 grid points in each direction, which implies that the mesh size is approximately 0.04 mm in each direction. The time step is set to \(7.8 \times 10^{-9}\) s.

The initial flames are obtained from direct numerical simulations of spherically expanding flames with CHEM1D. These initial flames each have a flame radius of approximately 0.85 mm at 1500 K with its centre located at the centre of the computational domain. Furthermore, all boundaries of the computational domain are modelled as outlet boundaries that are partially non-reflecting (cf. Section 4.3). The interaction of the flame with these boundaries cannot be accurately modelled in this manner. Therefore, the simulations are stopped before the flame reaches the boundaries of the domain, which in this particular case is after 0.8 ms of physical time.

The pressure of the initial flame is uniform in the domain. It takes approximately 3000 time steps (0.0234 ms of physical time) after the start of the simulation before the pressure field has established itself, and before other transient initial artefacts have left the computational domain.

As expected, the very small spherically expanding flames computed with the DNS program behave in a similar way as the larger ones simulated using the flamelet model in Chapter 2. This behaviour is shown in terms of the burning velocity, flame propagation velocity, gas velocity, and flame stretch rate throughout a spherically expanding flame in Fig. 4.5 for the DNS computations in which the chemistry is modelled with the single-step reaction mechanism and in Figs. 2.5a and 2.9a for the flamelet computations. The radius at the inner layer is approximately 1.17 mm (at ±1820 K) and 10.0 mm (at ±1640 K) in the DNS and flamelet computations, respectively. As discussed in Sections 2.5.2 and 2.7, these figures show that a smaller flame radius causes a stronger variation of the flame propa-
4.6 Validation of the DNS program

Figure 4.5: The burning velocity (solid line), flame propagation velocity (dashed line), gas velocity (dotted line), and flame stretch rate (dash-dotted line) throughout a spherically expanding flame computed with the DNS program in which the chemistry is modelled with the single-step reaction mechanism.

The variation in the flame propagation velocity throughout the flame. This variation indicates that the flame gets thinner while moving.

A comparison between the direct numerical simulations performed with the DNS program and CHEM1D is shown in Fig. 4.6, where the flame propagation velocity is plotted as a function of the flame radius, at 1500K. Fig. 4.6a shows that the behaviour of the flames computed with the two programs using the single-step reaction mechanism is very similar. However, a small quantitative disagreement of less than 1.5% is present between the DNS program and CHEM1D, with the former yielding slightly lower values. This small difference can be attributed to the distinctive assumptions and choices made in both programs. As mentioned in Section 4.6.1, the DNS program solves the full set of combustion equations, whereas CHEM1D uses the low-Mach number approximation. Furthermore, the DNS program uses a uniform grid with high-order spatial and temporal discretisation schemes, whereas CHEM1D uses an adaptive grid with less accurate discretisation schemes. However, the main part of the difference in the flame propagation velocity should be attributed to the boundary conditions. For the DNS program, these conditions are partially non-reflecting, which means that small pressure waves can enter the domain and disturb the flow field. This disturbance causes the flame to deviate increasingly from its spherical shape as the simulation proceeds. This behaviour is shown in Fig. 4.7a in which the mean flame radius and the difference between the maximum and minimum flame radius are plotted as a function of the flame propagation time, both at an isotherm of 1500K. However, the flame surface computed from this mean radius and the actual flame surface are almost identical as can be seen in Fig. 4.7b in which the flame surface is plotted as a function of the flame propagation time. This actual flame surface is calculated using an algorithm that has been developed by Geurts [35] and that has been proven to accurately determine an iso-surface of a scalar field. The nu-
Figure 4.6: The flame propagation velocity as a function of the flame radius at an isotherm of 1500 K for spherically expanding flames. The modelling of the chemistry is based on the single-step reaction mechanism (a) and on GRI-mech 3.0 (b). Solid line: CHEM1D with the single-step reaction mechanism, dashed line: the DNS program with the single-step reaction mechanism, dash-dotted line: the DNS program with the FGM method based on the single-step reaction mechanism (hardly visible), dot-dash-dotted line: CHEM1D with GRI-mech 3.0, dotted line: the DNS program with the FGM method based on GRI-mech 3.0.

Figure 4.7: The flame radius (a) and the flame surface (b) at an isotherm of 1500 K as a function of the flame propagation time. Solid line: mean flame radius, dashed line: difference between the maximum and minimum flame radius, dotted line: flame surface obtained from the mean flame radius, dash-dotted line: actual flame surface. Note that the dotted and dash-dotted lines in (b) coincide and that the chemistry is modelled with the single-step reaction mechanism.

Numerical data in Fig. 4.7 are obtained from simulations with the DNS program in which the chemistry is modelled with the single-step reaction mechanism.

Fig. 4.6a also shows the results of a flame simulation with the DNS program using the FGM method based on the single-step reaction mechanism. This simu-
4.7 TURBULENT FLAMES

The behaviour of an expanding flame in a turbulent flow field is dependent on both flame and flow field characteristics. This interaction is explained in more detail in Section 4.7.1. In Section 4.7.2, computational results are shown of expanding flames in different flow fields.

4.7.1 Combustion diagrams and regimes

The structure of premixed turbulent flames is dependent on the time and length scales of the turbulent flow and the chemical reactions. Based on these different scales, different combustion regimes can be identified in which the flames ex-
hibit very different behaviour. Borghi [13] was the first to classify these regimes by proposing a combustion diagram in terms of velocity and length scale ratios. Williams [115] proposed an equivalent combustion diagram in terms of the Reynolds and the Damköhler number. Several authors adjusted the combustion diagram proposed by Borghi, e.g., Abdel-Gayed et al. [1], Poinsot et al. [87, 88, 90], and Peters [82, 84, 86]. Abdel-Gayed et al. included the effect of Lewis numbers on the regimes based on experimental data. Poinsot et al. constructed the combustion diagram by using spectral diagrams obtained from DNS computations on the interaction between a vortex pair and a laminar flat flame. This slightly modifies the combustion diagram proposed by Borghi, especially in the prediction of flame quenching. Roberts et al. [94] constructed these spectral diagrams based on experiments. Peters introduced an extra regime called the thin reaction zones regime. The combustion diagram proposed by Peters is explained in more detail hereafter. Notice that the combustion diagrams are mainly based on essentially intuitive and dimensional considerations. Therefore, the diagrams can only be used qualitatively.

The combustion diagram as proposed by Peters [86] is shown in Fig. 4.9. The abscissa of this diagram is the ratio of the integral length scale $l_t$ and the flame thickness $\delta_f$, which represents a measure of the size of the turbulent eddies that interact with the flame. The ordinate is the ratio of the turbulent intensity $u'$ and the unstretched laminar burning velocity $s_0^l$, which is a measure of the velocity scales in the flow and the flame. In the construction of the diagram, it is assumed that all species have equal diffusion coefficients $D$ and that the Schmidt number $Sc = \nu/D$ is unity.

In the combustion diagram in Fig. 4.9, five different regimes can be identified: laminar flames, wrinkled flamelets, corrugated flamelets, thin reaction zones, and broken reaction zones. The most important aspects of these regimes are discussed hereafter.

Laminar flames are present if the turbulence Reynolds number is smaller than unity, otherwise the flames can be considered turbulent. The turbulence Reynolds number $Re_t$ is defined as the ratio of the turbulent inertial forces to the molecular viscous forces and can be rewritten in terms of the quantities along the axes of Fig. 4.9:

$$Re_t := \frac{u' l_t}{\nu} = \left( \frac{u'}{s_0^l} \right) \left( \frac{l_t}{\delta_f} \right). \quad (4.26)$$

The separation of the laminar regime from the turbulent regimes is indicated in Fig. 4.9 by the line where $Re_t = 1$.

Combustion in the flamelet regime occurs at short time scales and small length scales relative to the turbulence. The unburnt and burnt gases are separated from each other by a flame surface that has an inner structure similar to a laminar flame, and that is wrinkled or corrugated by the turbulent flow. In the wrinkled flamelet regime, the interaction between an initially flat flame and a turbulent flow can not lead to flame interactions. However, these interactions can occur in the corrugated flamelet regime. The separation between the wrinkled and the cor-
rugged flamelet regime is indicated in Fig. 4.9 by the line for which the turnover velocity \( u' \) of the integral scale eddies is equal to the laminar burning velocity \( s_0^L \).

The boundary of the flamelet regime to the thin reaction zones regime is given by the turbulence Karlovitz number \( \tilde{K}_t = 1 \), which is called the Klimov-Williams criterion. At this boundary the flame thickness \( \delta_f \) is equal to the size of the Kolmogorov scale eddies \( l_\eta \). This turbulence Karlovitz number is defined as the ratio between the chemical time scale \( \tau_c \) and the Kolmogorov time scale \( \tau_\eta \), and can be
rewritten in terms of the quantities along the axes of Fig. 4.9 (cf. [86]):

\[ K_{a_t} := \frac{\tau_c}{\tau_\eta} = \frac{\delta_f^2}{l_\eta^2} = \left( \frac{u'}{\delta_f} \right)^{3/2} \left( \frac{l_\eta}{\delta_f} \right)^{-1/2} \]  \hspace{1cm} (4.27)

In the thin reaction zones regime, the Kolmogorov scale eddies are smaller than the thickness of the flame: \( l_\eta < \delta_f \). Therefore, small eddies are able to enter and disturb the preheat zone, which is not possible in the flamelet regime. This is illustrated in Fig. 4.10. In the thin reaction zones regime, small eddies can enhance the transport of chemical species and heat in the preheat zone. However, these eddies are not able to influence the chemical reactions that take place at the inner layer of the flame, because the size of the Kolmogorov scale eddies \( l_\eta \) is larger than the thickness of the inner layer \( \delta_{il} \). Therefore, \( l_\eta = \delta_{il} \) represents the upper boundary of the thin reaction zones regime. This limit can also be written in terms of the turbulence Karlovitz number, because the thickness of the inner layer \( \delta_{il} \) can be related to the thickness of the flame \( \delta_f \) by a coefficient \( \zeta \) which is dependent on the pressure:

\[ \delta_{il} = \zeta \delta_f \] \hspace{1cm} (4.28)

Peters [83] shows that \( \zeta \) varies from \( \zeta = 0.1 \) at atmospheric pressure to \( \zeta = 0.03 \) at pressures around 30 times the atmospheric pressure. The upper boundary of the thin reaction zones regime can be written as:

\[ \zeta^2 K_{a_t} = 1 \] \hspace{1cm} (4.29)

Beyond the upper boundary of the thin reaction zones regime, the broken reaction zones regime is present. In this regime the Kolmogorov scale eddies are smaller than the thickness of the inner layer \( l_\eta < \delta_{il} \). Therefore, these eddies can locally influence the chemical reactions in the inner layer, thereby causing local extinction of the flame. Combustion in this regime is not considered here, because the local flame structure is neither continuous nor similar to laminar flame structures anymore.

### 4.7.2 Expanding flames

The developed DNS program is used here to simulate turbulent expanding flames. The first results of this evaluation are given in this section. Both the computational domain and the set of boundary conditions are chosen identical to those that are used in the flame simulations of the laminar spherically expanding flames shown in Section 4.6.2. This implies that the cubic computational domain has a length of 5.0 mm and 127 grid points in each direction, and that the boundaries are modelled as partially non-reflecting (cf. section 4.3). The time step is fixed at \( 7.8 \cdot 10^{-9} \) s. Furthermore, the chemistry is modelled with the single-step reaction mechanism (cf. Section 4.4.1). Simulations with the FGM method (cf. Section 4.4.2) give similar results.
As initial conditions, the laminar flame in Section 4.6.2 is used that is obtained from the DNS simulation using the single-step reaction mechanism. To ensure that this flame is not affected by any initial artefacts, its status at \( t = 0.156 \text{ ms} \) is implemented. This initial flame has a radius of 1.0mm at an isotherm of 1500K. It is superimposed onto an initial turbulent flow field, which is modelled to be homogeneous and isotropic. The decay of this field during the DNS calculations is negligible due to the very short simulated physical time frame.

This ambient initial turbulence field is chosen to satisfy certain specific requirements. A first one is that it should be divergence free to avoid the generation of pressure waves which could potentially affect the phenomena that are subject of the study. As a second requirement, the field needs to be sufficiently smooth to ensure that the governing transient equations can be solved with a minimum of discretisation errors. And finally, it is desirable to have a wave number range for which the turbulent cascade shows an inertial behaviour, as occurring in a homogeneous and isotropic turbulent flow field. In order to meet these conditions, the initial turbulence field is determined using a random number generation mechanism. In this implementation, the field’s stream function values on each grid point are defined by random numbers. Subsequently, this random field is convoluted with a three-dimensional top hat filter kernel of which the width is two times the mesh size. This kernel is approximated by a trapezoidal rule. It is applied repeatedly until the energy at the gridlevel is decreased sufficiently to be able to accurately determine the gradients in the DNS. In this particular case, it is repeated 125 times. Finally, this smoothed stream function is converted into a velocity field using the compact sixth-order discretisation scheme (cf. Section 4.2.1) to obtain a smooth velocity field. The resulting initial energy spectrum is equal to the initial spectrum of Ristorcelli and Blaisdell [93].

Three different initial turbulent flow fields are used here. Each field contains the same length scales \((l_t/\delta_f \approx 10)\) and different velocity scales \((u'/\delta_f \approx 0.9, 2.7, \text{ and } 9.1)\), i.e. the size and the position of the eddies in the computational domain are identical in each turbulent field but the turn-over velocities of these eddies are different. In the turbulent combustion diagram in Fig. 4.9, this corresponds to three positions on a vertical line. The length and velocity scales of the flow and the flame are chosen in such a way that these three positions are present at the combustion regimes that are of interest here: the wrinkled and corrugated flamelet regimes, as well as the thin reaction zones regime.

In Fig. 4.11, two-dimensional slices in the \( x - y \) plane through the centre of the computational domain are shown for the flame simulations with the various turbulent flow fields, at \( t = 0.25 \text{ ms} \). Also, the laminar flame that was used as initial condition is shown at this same moment in time. For each flame, three isotherms at \( T = 1000, 1400, \text{ and } 1800\text{K} \) are plotted.

The main characteristics of the different flame regimes that were discussed in Section 4.7.1 can be identified in these simulation results. In the thin reaction zones regime, the flame is heavily distorted and the flow field has caused the flame to be broadened at several positions along the flame due to eddies that were able to penetrate the preheat zone. However, in the flamelet regimes, the
Figure 4.11: Expanding flames in the laminar (a), wrinkled flamelets (b), corrugated flamelets (c), and thin reaction zones (d) regimes at the isotherms of 1000 K (dashed lines), 1400 K (solid lines), and 1800 K (dash-dotted lines).

global structure of the flame is still similar to the structure of the laminar flame. In the wrinkled flamelet regime, the flame shows the expected small distortion of the whole flame structure which is induced by the turbulent flow field. As expected, these distortions are more apparent in the corrugated flamelet regime.

4.8 Conclusions

A DNS program was developed and described in this chapter. Most of this kind of DNS programs have been developed to simulate flat flames in a turbulent flow
field. This program, however, was especially made to simulate expanding flames in a turbulent environment. Therefore, all boundaries are considered to be outlet boundaries. They were implemented as partially non-reflecting boundaries, which enables the gases and the acoustic waves to leave the computational domain. The computational costs of DNS are very high due to the large and stiff set of combustion equations that have to be solved. This computational effort is commonly reduced by using a simplified chemical reaction mechanism. Besides the implementation of a single-step reaction mechanism, the recently developed Flamelet-Generated Manifold (FGM) method was also implemented. Both methods showed a very large reduction in computational time and were shown to give accurate results for simulations with unit Lewis numbers.

The DNS program was validated by simulating two kinds of laminar flames: flat stretchless flames and spherically expanding flames. These flat flames were shown to be similar to flames computed with a numerical one-dimensional program that had been proven to give very accurate results. The spherically expanding flames were also accurately reproduced. Further calculations were performed on expanding flames in turbulent flow fields with various intensities. It appeared that these various fields influence the behaviour of the expanding flames. Overall, it can be concluded that the DNS program is well suitable for the simulation of expanding flames in a turbulent flow field.
DIRECT NUMERICAL SIMULATIONS
Conclusions and recommendations

The underlying investigation can be subdivided into three parts. First, a new flamelet model was introduced for the simulation of perfectly spherical and cylindrical flames. This flamelet model was used to investigate the behaviour of these flames. Then, the relation between the mass burning rate and flame stretch was analysed in detail. As a final topic, a Direct Numerical Simulation (DNS) program was developed for simulating expanding flames in a turbulent flow field. The most important aspects and results of each of these subjects are summarized below together with some recommendations for future research.

In Chapter 2, a new flamelet model was developed. This model was shown to very accurately simulate the perfectly spherical and cylindrical flames without being computationally expensive. With this model, simulations were performed on stoichiometric methane-air flames under atmospheric pressure. The analyses of various combustion variables revealed some interesting new insights, such as the fact that the mass burning rate at the inner layer of steady spherical and cylindrical flames is almost independent of the flame radius. In future research, the flamelet model can be used to simulate and investigate different kinds of spherical and cylindrical flames in various conditions, e.g. lean and rich flames, smaller and larger flame radii, lower and higher pressures, and other gas mixtures.

In Chapter 3, the relation between the mass burning rate and the flame stretch rate was analysed by deducing expressions for the Markstein number. An expression for the Markstein number that takes the inner structure of a flame into account was derived. It was shown that this expression can predict the Markstein number more accurately than existing expressions that assume an infinitely thin reaction layer. However, it is still difficult to accurately predict the effects of preferential diffusion on the Markstein number. This is an interesting topic for future research. In Chapter 3, it is also shown that it is not useful to use two Markstein numbers: one that takes the curvature of the flame into account and another one that incorporates the straining of the flow, because the Markstein number for the flow straining is not uniquely defined, i.e. this number has different values in different combustion situations.

In Chapter 4, a DNS program was developed to simulate freely expanding turbulent flames. Besides the simulation of expanding flames instead of flat flames,
two other aspects of this program are different from most existing DNS programs: the modelling of all boundaries as outlet boundaries to prevent pressure build-up, and the use of the Flamelet-Generated Manifold (FGM) method to reduce the computational costs. This program was successfully validated in the simulation of flat stretchless flames and perfectly spherical flames. First simulations of turbulent expanding flames and first analyses were performed. These simulations showed that the DNS program is suitable for further research on freely expanding turbulent flames. However, the use of the FGM method in turbulent flame simulations should be studied in more detail. Furthermore, it would be interesting to investigate if preferential diffusion effects can be included by increasing the dimensions of the manifold in the FGM method. In spite of the increase in computational costs, it is recommended to use larger computational domains for the simulations to prevent the flame from reaching the computational boundaries, because both the flame and the turbulent flow field at the boundaries are influenced by these boundaries. The resulting turbulent flames can be used to study various interesting aspects of these flames, such as the influence of the flame curvature, flow straining, and different turbulent flow fields.

Overall, it can be concluded that the flamelet model and DNS program are able to accurately simulate perfectly spherical and cylindrical flames and turbulent expanding flames, respectively. Furthermore, the analyses of the spherical and cylindrical flames have increased our understanding of the general behaviour of these flames.
A

Derivation of an expression for $m_{il}^0$

In this appendix, an expression for the stretchless mass burning rate at the inner layer $m_{il}^0$ is derived by applying Integral Analysis (cf. [24]) to Eq. (2.12) and the flamelet equation for the scalar field $Y$:

$$\frac{\partial}{\partial s} (\sigma m Y) - \frac{\partial}{\partial s} \left( \sigma \frac{\lambda}{Le y_c_p} \frac{\partial Y}{\partial s} \right) - \sigma \rho_Y = -\sigma \rho K_Y , \quad (A.1)$$

in which $Y$ can represent the enthalpy or one of the species mass fractions that obeys $|\nabla Y| \neq 0$. This flamelet equation (A.1) is derived from the conservation equation of $Y$:

$$\frac{\partial (\rho Y)}{\partial t} + \nabla \cdot (\rho v Y) - \frac{1}{Le y} \nabla \cdot \left( \frac{\lambda}{c_p} \nabla Y \right) = \rho Y , \quad (A.2)$$

in the same way as the derivation of the other flamelet equations in Section 2.2.3.

For convenience, Eq. (A.1) is written in terms of:

$$C(s) := \frac{\partial}{\partial s} \left( \sigma m^0 Y^0 \right) , \quad (A.3)$$

$$\mathcal{F}(s) := \sigma \frac{\lambda}{Le y_c_p} \frac{\partial Y^0}{\partial s} , \quad (A.4)$$

$$\mathcal{R}(s) := \sigma \rho_Y , \quad (A.5)$$

and for stretchless flames, which means that $K = 0$:

$$C(s) - \frac{\partial}{\partial s} \mathcal{F}(s) - \mathcal{R}(s) = 0 . \quad (A.6)$$

Eq. (A.6) is multiplied with $\mathcal{F}$ and subsequently integrated over $s$ from the unburnt to the burnt side of the flame:

$$\int_{s_u}^{s_b} C(s) \mathcal{F}(s) \, ds = \int_{s_u}^{s_b} \mathcal{R}(s) \mathcal{F}(s) \, ds + \frac{1}{2} \left( \mathcal{F}(s_b)^2 - \mathcal{F}(s_u)^2 \right) . \quad (A.7)$$
The last term on the right-hand side of Eq. (A.7) is zero, because $\partial Y_0 / \partial s$ and, therefore, $F$ is zero at the unburnt and the burnt side of the flame.

An expression for $F$ in the left-hand side of Eq. (A.7) is obtained, if Eq. (A.6) is integrated over the preheat zone:

$$ F(s) = \int_{s_u}^{s_b} (C(s) - R(s)) \, ds. \quad (A.8) $$

In the preheat zone, the reactive term is very small compared to the convective term: $|R(s)| \ll |C(s)|$. Therefore, Eq. (A.8) reduces to

$$ F(s) \approx \int_{s_u}^{s_b} C(s) \, ds. \quad (A.9) $$

This approximation is valid if the reaction zone is much thinner than the preheat zone, which is the case in the flames considered here.

Substitution of Eq. (A.9) into the left-hand side of Eq. (A.7) and elaboration of the resulting integral gives:

$$ \frac{1}{2} \left( \int_{s_u}^{s_b} C(s) \, ds \right)^2 \approx \int_{s_u}^{s_b} R(s) F(s) \, ds. \quad (A.10) $$

Using Eq. (A.3), the integral in the left-hand side of Eq. (A.10) becomes:

$$ \int_{s_u}^{s_b} C(s) \, ds = \left( \sigma m^0 \right)_l \left( Y_0^b - Y_u \right). \quad (A.11) $$

In obtaining Eq. (A.11), $\sigma m^0 = \text{constant}$ is used, which can be deduced from integration of Eq. (2.12) with $K = 0$. Using Eqs. (A.4) and (A.5), the right-hand side of Eq. (A.10) results in:

$$ \int_{s_u}^{s_b} R(s) F(s) \, ds = \int_{Y_u}^{Y_0^b} \lambda \frac{\rho_y}{Le \gamma c_p} \, dY. \quad (A.12) $$

By substituting Eqs. (A.11) and (A.12) into Eq. (A.10), the expression of the stretchless mass burning rate at the inner layer in Eq. (2.23) is obtained:

$$ m_{il}^0 \approx \frac{1}{|Y_0^b - Y_u|} \sqrt{\frac{2}{\int_{Y_u}^{Y_0^b} \left( \frac{\sigma}{\sigma_l} \right)^2 \frac{\lambda}{Le \gamma c_p} \rho_y \, dY}}. \quad (A.13) $$
## Derivation of expressions for the Markstein number

The dependence of the mass burning rate \( m_0^b \) on the enthalpy \( h_0^b \) and the mole number \( \psi_{j,b}^0 \) in Eq. (3.5) is discussed for lean hydrocarbon-air flames in Section B.1. This dependence has to be known in order to derive the expressions (3.14) and (3.20) for the Markstein number. These expressions are derived in Sections B.2 and B.3, respectively.

### B.1 Dependence of \( m_0^b \) on \( h_0^b \) and \( \psi_{j,b}^0 \)

For convenience, Eq. (3.5) can be rewritten as:

\[
\frac{m_b}{m_b^0} = 1 - IK_T^0
- \sum_{i=1}^{N_s} \left( \frac{IK_0^0}{Le_i} - IK_T^0 \right) \left( \phi_{i,b}^0 - \phi_{i,u} \right) \left( \mu_{ji} \cdot \nabla \psi \ln \left( m_0^b \right) \right) + \text{hot},
\]  

(B.1)

with the vectors

\[
\mu_{ji} := (\mu_{C_i}, \mu_{H_i}, \mu_{O_i}, M_i h_i^*)^T
\]  

(B.2)

and

\[
\nabla \psi := \left( \frac{\partial}{\partial \psi_{C_i,b}}, \frac{\partial}{\partial \psi_{H_i,b}}, \frac{\partial}{\partial \psi_{O_i,b}}, \frac{\partial}{\partial h_0^b} \right)^T.
\]  

(B.3)

The Karlovitz integrals \( IK_0^0 \) and \( IK_T^0 \) in Eq. (B.1) are defined in Eqs. (3.8) and (3.9), respectively:

\[
IK_i^0 := \frac{Le_i}{(\sigma m_i^0)^b} \int_{s_u}^{s_b} \sigma \rho K \psi_i^0 ds
\]

(B.4)
and

\[ H_k^0 := \frac{1}{(\sigma m^0)_b} \int_a^b \alpha \rho^0 k T^0 \, ds. \]  

(B.5)

The term \( \mu_{ij} \cdot \nabla \psi \ln(m^0) \) in Eq. (B.1) is difficult to obtain, because the explicit dependence of the mass burning rate \( m^0 \) on the enthalpy \( k^0 \) and the mole number \( \psi^0_{i,b} \) is not known theoretically. However, expressions for the mass burning rate \( m^0 \) in terms of the flame temperature \( T^0_b \) and some product mole numbers \( \psi^0_{i,b} \) are given more frequently in literature. The partial derivatives \( \partial \ln (m^0) / \partial \psi^0_{i,b} \) and \( \partial \ln (m^0) / \partial m^0 \) in Eq. (B.1) can only be rewritten in terms of the product mole numbers \( \psi^0_{i,b} \) and the flame temperature \( T^0_b \), if a one-to-one relation exists between the set of variables \( \Psi := (\psi^0_{C,b}, \psi^0_{H,b}, \psi^0_{O,b}, T^0_b) \) on the one hand and \( \Phi := (\phi^0_{CO_2,b}, \phi^0_{H_2O,b}, \phi^0_{O_2,b}, T^0_b) \) on the other. In case of lean methane-air flames with five species \( \text{CH}_4, \text{O}_2, \text{H}_2\text{O}, \text{CO}_2, \) and \( \text{N}_2 \) and four elements \( \text{C}, \text{O}, \text{H}, \) and \( \text{N} \), a simple relationship exists, because, at the equilibrium state, no radicals are present and the mole number of methane is zero. Furthermore, nitrogen can be treated as an inert gas. Therefore, for the burnt gases of these lean flames, the following relations hold:

\[
\begin{align*}
F_1(\Psi, \Phi) &:= \hat{\psi}^0_{C,b} - \hat{\phi}^0_{CO_2,b} = 0, \\
F_2(\Psi, \Phi) &:= \hat{\psi}^0_{H,b} - 2\hat{\phi}^0_{H_2O,b} = 0, \\
F_3(\Psi, \Phi) &:= \hat{\psi}^0_{O,b} - 2\hat{\phi}^0_{CO_2,b} - \hat{\phi}^0_{H_2O,b} - 2\hat{\phi}^0_{O_2,b} = 0, \\
F_4(\Psi, \Phi) &:= \hat{\phi}^0 - c_p (\hat{\theta}^0 - \hat{\theta}_{\text{ref}}) \\
&\quad - \hat{h}_{\text{CO}_2} \hat{\phi}^0_{CO_2,b} - \hat{h}_{\text{H}_2\text{O}} \hat{\phi}^0_{H_2O,b} - \hat{h}_{\text{O}_2} \hat{\phi}^0_{O_2,b} = 0,
\end{align*}
\]

(B.6)

where \( \hat{\theta}_{\text{ref}} \) is the dimensionless temperature at which the formation enthalpies \( \hat{h}_{\text{CO}_2}, \hat{h}_{\text{H}_2\text{O}}, \) and \( \hat{h}_{\text{O}_2} \) are defined. In the set of equations (B.6), the following scaled dimensionless quantities

\[
\hat{\psi}_j := \frac{\psi^0_j}{\phi_{\text{CH}_4,u}}, \quad \hat{\phi}_i := \frac{\phi^0_i}{\phi_{\text{CH}_4,u}}, \quad \hat{\theta}^0 := \frac{c_p T^0}{M_{\text{CH}_4} h^*_\text{CH}_4}, \\
\hat{h}^0 := \frac{h^0}{M_{\text{CH}_4} h^*_\text{CH}_4}, \quad \hat{h}^*_i := \frac{M_i h^*_i}{M_{\text{CH}_4} h^*_\text{CH}_4},
\]

(B.7)

and the vectors \( \Psi := (\hat{\psi}^0_{C,b}, \hat{\psi}^0_{H,b}, \hat{\psi}^0_{O,b}, \hat{\phi}^0) \) and \( \Phi := (\hat{\phi}^0_{CO_2,b}, \hat{\phi}^0_{H_2O,b}, \hat{\phi}^0_{O_2,b}, \hat{\phi}^0) \) are used. From the implicit function theorem, this set of equations (B.6)

\[ F(\Psi, \Phi) := (F_1(\Psi, \Phi), \ldots, F_4(\Psi, \Phi))^T = 0 \]

(B.8)
implicitly defines $\hat{\Phi}$ as a function of $\hat{\Psi}$, provided that $J_{\Phi} = \partial F / \partial \Phi$ exists. For $J_{\Phi}$, from the set of equations (B.6) it can be found that:

$$J_{\Phi} = -\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 2 & 0 & 0 \\ 2 & 1 & 2 & 0 \\ \hat{h}_{CO_2}^r & \hat{h}_{H_2O}^r & \hat{h}_{O_2}^r & 1 \end{pmatrix}. \quad \text{(B.9)}$$

Furthermore, the matrix $V = \partial \hat{\Phi} / \partial \hat{\Psi}$ can be computed from $J_{\Phi} V = -J_{\Psi}$, with $J_{\Psi} = \partial F / \partial \Psi = I$, the unit matrix, so that $V$ is given by

$$V = -J_{\Phi}^{-1} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \frac{1}{2} & 0 & 0 \\ -1 & 0 & 0 & 0 \\ (\hat{h}_{O_2}^r - \hat{h}_{CO_2}^r) & (\frac{1}{2}\hat{h}_{O_2}^r - \frac{1}{2}\hat{h}_{H_2O}^r) & -\frac{1}{2}\hat{h}_{O_2}^r & 1 \end{pmatrix}. \quad \text{(B.10)}$$

Subsequently, it is possible to rewrite the derivative of the mass burning rate $m_b^0$ in terms of $\Phi$ using the chain rule:

$$\mu_{ji} \cdot \nabla \Phi \ln \left( m_b^0 \right) = (\nabla \mu_{ji}) \cdot \nabla \Phi \ln \left( m_b^0 \right). \quad \text{(B.11)}$$

### B.2 $M_b$ for an infinitely thin reaction layer

An expression for the Markstein number of weakly stretched flames is derived using integral analysis. It is assumed that the reaction layer is infinitely thin. The reaction layer is then positioned at $s_b = 0$ and the preheat zone extends from $s_u = -\infty$ to $s_b = 0$. For weakly stretched flames, the flame stretch rate becomes uniform throughout the flame $K(s) = K_b$. Furthermore, the contributions of the higher-order terms in the Karlovitz integrals (B.4) and (B.5) are neglected. The heat conductivity $\lambda$ and the heat capacity at constant pressure $c_p$ are assumed to be independent of temperature. From convective-diffusive balances, it can be found that the structure of the undistorted temperature and mole number profiles in the preheat zone can be written as:

$$\tilde{T}^0(s) = \exp \left( \frac{s}{\delta_f^0} \right) \quad \text{(B.12)}$$

and

$$\tilde{\phi}_i^0(s) = \exp \left( \frac{Le_i s}{\delta_f^0} \right), \quad \text{(B.13)}$$

respectively. Furthermore, the flame surface area $\sigma$ is equal to one and the density is simply related to the temperature by

$$\rho^0(s) = \frac{\rho_u \tilde{T}_u}{\tilde{T}^0(s)} = \frac{\rho_u}{1 + \tau \exp \left( \frac{s}{\delta_f^0} \right)} \quad \text{.}$$

(B.14)
After substitution of Eqs. (B.12) and (B.14) into Eq. (B.5), the Karlovitz integral $IK_0^T$ reduces to:

$$IK_0^T = \frac{\rho_0 K_b}{m_b^0} \int_{-\infty}^{0} \frac{\exp \left( \frac{s}{\delta_f^0} \right)}{1 + \tau \exp \left( \frac{s}{\delta_f^0} \right)} \, ds$$

$$= K_{ab} \left( \frac{1 + \tau}{\tau} \right) \ln (1 + \tau), \quad (B.15)$$

with $K_{ab} := \rho_0^0 K_b \delta_f^0 / m_b^0$. The Karlovitz integral $IK_0^i$ in Eq. (B.4) can be derived in a similar way by substituting Eqs. (B.13) and (B.14) into Eq. (B.4). $IK_0^i / Le_i$ then becomes:

$$IK_0^i = \frac{\rho_0 K_b}{m_b^0} \int_{-\infty}^{0} \frac{\exp \left( \frac{Le_i s}{\delta_f^0} \right)}{1 + \tau \exp \left( \frac{s}{\delta_f^0} \right)} \, ds$$

$$= K_{ab} (1 + \tau) \tau^{-Le_i} \int_{0}^{\tau} \frac{y^{Le_i-1}}{1 + y} \, dy \quad (B.16)$$

This integral can be rewritten using partial integration. Subtraction of Eq. (B.16) from Eq. (B.15) results in:

$$\left( \frac{IK_0^i}{Le_i} - IK_0^T \right) = K_{ab} (1 + \tau) (1 - Le_i) F_i, \quad (B.17)$$

with

$$F_i = \int_{0}^{\tau} \ln \left( \frac{1 + y}{y^2} \right) \left( \frac{y}{\tau} \right)^{Le_i} \, dy \quad (B.18)$$

Substituting Eq. (B.17) into Eq. (3.5) and combining it with Eq. (3.4) gives the following expression for the Markstein number:

$$M_b = \left( \frac{1 + \tau}{\tau} \right) \ln (1 + \tau)$$

$$+ (1 + \tau) \sum_{i=1}^{N_i} F_i (1 - Le_i) \left( \hat{\phi}_{i,b} - \hat{\phi}_{i,a} \right) \left( \mu_{ji} \cdot \nabla \psi \ln \left( m_b^0 \right) \right), \quad (B.19)$$

in which the scaled dimensionless quantities defined in Eq. (B.7) are used and in which the higher-order terms in Eqs. (3.4) and (3.5) are neglected. Using Eqs. (B.10) and (B.11) to compute the partial derivatives of $m_b^0$ with respect to $h_b^0$ and $\psi_{i,b}$.
Eq. (B.19) becomes:

$$M_b = \left(1 + \frac{\tau}{\tau}\right) \ln (1 + \tau) + (1 + \tau) \times$$

$$\left(( F_{\text{CH}_4} L_{\text{CH}_4} - F_{\text{CO}_2} L_{\text{CO}_2} + F_{\text{CO}_2} - F_{\text{CH}_4}) \frac{\partial \ln (m^0_b)}{\partial \phi_{\text{CO}_2,b}} \right. \left. + 2 \left( F_{\text{CH}_4} L_{\text{CH}_4} - F_{\text{H}_2\text{O}} L_{\text{H}_2\text{O}} + F_{\text{H}_2\text{O}} - F_{\text{CH}_4}\right) \frac{\partial \ln (m^0_b)}{\partial \phi_{\text{H}_2\text{O},b}} \right)$$

$$+ 2 \left( F_{\text{O}_2} L_{\text{O}_2} - F_{\text{CH}_4} L_{\text{CH}_4} + F_{\text{CH}_4} - F_{\text{O}_2}\right) \frac{\partial \ln (m^0_b)}{\partial \phi_{\text{O}_2,b}}$$

$$+ F_{\text{CH}_4} (L_{\text{CH}_4} - 1) \left( \partial^{0}_{b} - \hat{\theta}_u \right) \frac{\partial \ln (m^0_b)}{\partial \phi^{0}_{b}} \right).$$

(B.20)

In the derivation of Eq. (B.20), the following substitutions were made:

$$\hat{\phi}_{\text{CO}_2,b} = \hat{\phi}_{\text{CH}_4,u} = 1,$$

$$\hat{\phi}_{\text{CO}_2,u} = \hat{\phi}_{\text{H}_2\text{O},u} = 0,$$

$$\hat{\phi}_{\text{H}_2\text{O},b} = 2,$$

$$\hat{\phi}_{\text{O}_2,b} - \hat{\phi}_{\text{O}_2,u} = -2.$$

(B.21)

In case it is assumed that $m^0_b$ depends only explicitly on $T^0_b$, Eq. (B.20) for the Markstein number reduces to:

$$M_b = \left(1 + \frac{\tau}{\tau}\right) \ln (1 + \tau)$$

$$+ (1 + \tau) F_{\text{fuel}} \left(L_{\text{fuel}} - 1\right) \left( \partial^{0}_{b} - \hat{\theta}_u \right) \frac{\partial \ln (m^0_b)}{\partial \phi^{0}_{b}}.$$

(B.22)

Notice that Eq. (B.22) is not only valid for methane-air flames, but for lean hydrocarbon-air flames in general. This expression for the Markstein number is shown in Eq. (3.14) for unscaled parameters.

**B.3 $M_{il}$ for a finite thickness of the reaction layer**

For weakly stretched flames with a finite thickness of the reaction layer, an expression for the Markstein number at the inner layer $M_{il}$ can be obtained from
the combination of Eqs. (2.12), (3.4), and (3.5):

$$M_{il} = \frac{1}{\delta^0_{\rho_{il}}^0} \left( I_T - \int_{s_d}^{s_h} \rho^0 \, ds \right) \left( \phi_{0,il} - \phi_{0,i} \right) \left( \mu_{ji} \cdot \nabla \psi \ln \left( m_b^0 \right) \right),$$

(B.23)

with the integrals

$$I_T := \int_{s_u}^{s_h} \rho^0 \hat{s}^0 \, ds,$$

(B.24)

and

$$I_i := \int_{s_u}^{s_h} \rho^0 \phi^0_i \, ds,$$

(B.25)

and with the vectors $\mu_{ji}$ and $\nabla \psi$ defined in Eqs. (B.2) and (B.3), respectively. In the derivation of Eq. (B.23), the higher-order terms in Eqs. (3.4) and (3.5) are neglected and it is assumed that the flame stretch rate becomes uniform throughout the flame for weakly stretched flames. Eq. (B.23) for the Markstein number can be rewritten in terms of $\partial m_b^0 / \partial \phi_{0,b}^0$ and $\partial m_b^0 / \partial T_b^0$ instead of $\partial m_b^0 / \partial \phi_{1,b}^0$ and $\partial m_b^0 / \partial T_b^0$, by substituting Eqs. (B.10), (B.11), and (B.21) into Eq. (B.23):

$$M_{il} = \frac{1}{\delta^0_{\rho_{il}}^0} \left( I_T - \int_{s_d}^{s_h} \rho^0 \, ds \right) \frac{\partial \ln (m_b^0)}{\partial \phi_{CO_2,b}^0} (I_{CO_2} - I_{CH_4})$$

$$+ 2 \frac{\partial \ln (m_b^0)}{\partial \phi_{H_2O,b}^0} (I_{H_2O} - I_{CH_4}) + 2 \frac{\partial \ln (m_b^0)}{\partial \phi_{O_2,b}^0} (I_{CH_4} - I_{O_2})$$

$$- (\hat{\phi}_b^0 - \hat{\theta}_a) \frac{\partial \ln (m_b^0)}{\partial \phi_b^0} (I_{CH_4} - I_T),$$

(B.26)

thereby using the dimensionless quantities defined in Eq. (B.7). If it is assumed that $m_b^0$ depends only explicitly on $T_b^0$, the following expression for the Markstein number can be derived that is not only valid for lean methane-air flames, but for lean hydrocarbon-air flames in general:

$$M_{il} = \frac{1}{\delta^0_{\rho_{il}}^0} \left( I_T - \int_{s_d}^{s_h} \rho^0 \, ds \right) (\hat{\theta}_b^0 - \hat{\theta}_a) \frac{\partial \ln (m_b^0)}{\partial \phi_b^0} (I_{fuel} - I_T).$$

(B.27)

This expression for the Markstein number is given in Eq. (3.20) for unscaled parameters.
Nomenclature

Roman characters

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
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<tr>
<td>$A_i$</td>
<td>chemical symbol for species $i$</td>
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<tr>
<td>$A$</td>
<td>area</td>
<td>m$^2$</td>
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<tr>
<td>$A_c$</td>
<td>pre-exponential constant</td>
<td>$^*$</td>
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<td>$a$</td>
<td>applied strain rate</td>
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<td>$C_1$</td>
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<td>kg m$^{-1}$ s$^{-1}$ K$^{-1/2}$</td>
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<tr>
<td>$C_2$</td>
<td>constant in Sutherland’s theory of viscosity</td>
<td>K</td>
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<td>$C_r, C_0$</td>
<td>constants related to the flame radius</td>
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<tr>
<td>$c$</td>
<td>travelling velocity of acoustic waves</td>
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<td>$c_p$</td>
<td>heat capacity at constant pressure</td>
<td>J kg$^{-1}$ K$^{-1}$</td>
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<td>binary diffusion coefficient</td>
<td>m$^2$ s$^{-1}$</td>
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<tr>
<td>$D$</td>
<td>diffusion coefficient</td>
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<td>mixture-averaged diffusion coefficient</td>
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<td>scalar field describing the flame shape</td>
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<td>$I_K$</td>
<td>Karlovitz integral</td>
<td>–</td>
</tr>
<tr>
<td>$K_c$</td>
<td>equilibrium constant</td>
<td>–</td>
</tr>
<tr>
<td>$K$</td>
<td>flame stretch rate</td>
<td>s$^{-1}$</td>
</tr>
<tr>
<td>$K_p$</td>
<td>pressure relaxation constant</td>
<td>s$^{-1}$</td>
</tr>
<tr>
<td>$k$</td>
<td>reaction rate coefficient</td>
<td>$^*$</td>
</tr>
<tr>
<td>$\mathcal{L}$</td>
<td>Markstein length</td>
<td>m</td>
</tr>
<tr>
<td>$L$</td>
<td>characteristic length of the domain</td>
<td>m</td>
</tr>
</tbody>
</table>
### Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Meaning</th>
</tr>
</thead>
<tbody>
<tr>
<td>$L_{1-N_f+5}$</td>
<td>amplitudes of the characteristic waves</td>
</tr>
<tr>
<td>$l_t$</td>
<td>integral length scale</td>
</tr>
<tr>
<td>$l_\eta$</td>
<td>Kolmogorov length scale</td>
</tr>
<tr>
<td>$M$</td>
<td>mass</td>
</tr>
<tr>
<td>$M_i$</td>
<td>species molar mass</td>
</tr>
<tr>
<td>$\bar{M}$</td>
<td>mean molar mass</td>
</tr>
<tr>
<td>$m$</td>
<td>mass burning rate</td>
</tr>
<tr>
<td>$N_e$</td>
<td>number of elements</td>
</tr>
<tr>
<td>$N_r$</td>
<td>number of reactions</td>
</tr>
<tr>
<td>$N_s$</td>
<td>number of species</td>
</tr>
<tr>
<td>$n$</td>
<td>molar concentration</td>
</tr>
<tr>
<td>$n_f$</td>
<td>unit vector normal to the flame surface</td>
</tr>
<tr>
<td>$\mathcal{P}$</td>
<td>stress tensor</td>
</tr>
<tr>
<td>$p$</td>
<td>hydrostatic pressure</td>
</tr>
<tr>
<td>$p_{at}$</td>
<td>atmospheric pressure</td>
</tr>
<tr>
<td>$Q$</td>
<td>reaction rate</td>
</tr>
<tr>
<td>$q$</td>
<td>heat flux field</td>
</tr>
<tr>
<td>$R$</td>
<td>universal gas constant</td>
</tr>
<tr>
<td>$R_f$</td>
<td>radius of the flame front</td>
</tr>
<tr>
<td>$r$</td>
<td>radius</td>
</tr>
<tr>
<td>$r, \theta, \varphi$</td>
<td>spherical polar coordinates</td>
</tr>
<tr>
<td>$r, \theta, z$</td>
<td>cylindrical polar coordinates</td>
</tr>
<tr>
<td>$s_T$</td>
<td>source term of the temperature</td>
</tr>
<tr>
<td>$s_{Y_j}$</td>
<td>source term of the mass fraction $Y_j$</td>
</tr>
<tr>
<td>$s$</td>
<td>coordinate perpendicular to the flame surface</td>
</tr>
<tr>
<td>$s_L$</td>
<td>laminar burning velocity</td>
</tr>
<tr>
<td>$T$</td>
<td>temperature</td>
</tr>
<tr>
<td>$T^*$</td>
<td>reference temperature</td>
</tr>
<tr>
<td>$t$</td>
<td>time</td>
</tr>
<tr>
<td>$u, v, w$</td>
<td>velocity components in cartesian coordinate system</td>
</tr>
<tr>
<td>$u'$</td>
<td>turbulent intensity</td>
</tr>
<tr>
<td>$\bar{u}$</td>
<td>internal energy density</td>
</tr>
<tr>
<td>$V$</td>
<td>diffusion velocity field</td>
</tr>
<tr>
<td>$V$</td>
<td>volume</td>
</tr>
<tr>
<td>$v$</td>
<td>velocity field</td>
</tr>
<tr>
<td>$v_f$</td>
<td>flame propagation velocity field</td>
</tr>
<tr>
<td>$X_i$</td>
<td>species mol fraction</td>
</tr>
<tr>
<td>$x, y, z$</td>
<td>cartesian coordinates</td>
</tr>
<tr>
<td>$Y$</td>
<td>scalar field describing the flame shape</td>
</tr>
<tr>
<td>$Y_i$</td>
<td>species mass fraction</td>
</tr>
</tbody>
</table>

* The unit depends on the situation.
**NOMENCLATURE**

Greek characters

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha$</td>
<td>reaction order</td>
<td>–</td>
</tr>
<tr>
<td>$\alpha_{1.2}$</td>
<td>discretisation coefficients</td>
<td>–</td>
</tr>
<tr>
<td>$\beta$</td>
<td>temperature exponent</td>
<td>–</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>specific heat ratio</td>
<td>–</td>
</tr>
<tr>
<td>$\delta_f$</td>
<td>flame thickness</td>
<td>m</td>
</tr>
<tr>
<td>$\delta_{il}$</td>
<td>inner layer thickness</td>
<td>m</td>
</tr>
<tr>
<td>$\epsilon$</td>
<td>relative error</td>
<td>–</td>
</tr>
<tr>
<td>$\epsilon$</td>
<td>perturbation parameter</td>
<td>–</td>
</tr>
<tr>
<td>$\zeta$</td>
<td>thickness coefficient</td>
<td>–</td>
</tr>
<tr>
<td>$\eta$</td>
<td>flame-adapted coordinate system</td>
<td>m</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>heat conductivity</td>
<td>J K$^{-1}$ m$^{-1}$ s$^{-1}$</td>
</tr>
<tr>
<td>$\lambda_{1-N_{i+5}}$</td>
<td>characteristic velocities</td>
<td>m s$^{-1}$</td>
</tr>
<tr>
<td>$\mu$</td>
<td>dynamic viscosity</td>
<td>kg m$^{-1}$ s$^{-1}$</td>
</tr>
<tr>
<td>$\mu_{ji}$</td>
<td>species composition</td>
<td>–</td>
</tr>
<tr>
<td>$\nu$</td>
<td>kinematic viscosity</td>
<td>m$^2$ s$^{-1}$</td>
</tr>
<tr>
<td>$\gamma_{li}$</td>
<td>molar stoichiometric coefficient</td>
<td>–</td>
</tr>
<tr>
<td>$\xi, \zeta, \eta$</td>
<td>flame-adapted coordinates</td>
<td>m</td>
</tr>
<tr>
<td>$\rho$</td>
<td>mass density</td>
<td>kg m$^{-3}$</td>
</tr>
<tr>
<td>$\dot{\rho}_i$</td>
<td>chemical source term</td>
<td>kg m$^{-3}$ s$^{-1}$</td>
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<tr>
<td>$\sigma$</td>
<td>area field</td>
<td>m$^2$</td>
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<tr>
<td>$\tau$</td>
<td>viscous stress tensor</td>
<td>kg m$^{-1}$ s$^{-2}$</td>
</tr>
<tr>
<td>$\tau_c$</td>
<td>chemical time scale</td>
<td>s</td>
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<tr>
<td>$\tau_{\eta}$</td>
<td>Kolmogorov time scale</td>
<td>s</td>
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<tr>
<td>$\phi$</td>
<td>equivalence ratio</td>
<td>–</td>
</tr>
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<td>$\phi_i$</td>
<td>species mole number</td>
<td>mol kg$^{-1}$</td>
</tr>
<tr>
<td>$\psi_j$</td>
<td>element mole number</td>
<td>mol kg$^{-1}$</td>
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</table>

Subscripts

<table>
<thead>
<tr>
<th>Symbol</th>
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</thead>
<tbody>
<tr>
<td>$A$</td>
<td>area based</td>
</tr>
<tr>
<td>$ad$</td>
<td>adiabatic</td>
</tr>
<tr>
<td>$b$</td>
<td>burnt</td>
</tr>
<tr>
<td>$c$</td>
<td>curvature</td>
</tr>
<tr>
<td>$i$</td>
<td>species index</td>
</tr>
<tr>
<td>$il$</td>
<td>inner layer</td>
</tr>
<tr>
<td>$j$</td>
<td>element index</td>
</tr>
<tr>
<td>$k$</td>
<td>grid point index</td>
</tr>
<tr>
<td>$l$</td>
<td>reaction index</td>
</tr>
<tr>
<td>$M$</td>
<td>mass based</td>
</tr>
<tr>
<td>$m$</td>
<td>mixture</td>
</tr>
<tr>
<td>$p$</td>
<td>species index</td>
</tr>
</tbody>
</table>
Nomenclature

$s$ strain
$u$ unburnt

**Superscripts**

- 0 unstretched
- $b$ backward
- $f$ forward
- $m$ time index
- $n$ normal direction
- $r$ radial direction
- $t$ tangential direction

**Dimensionless numbers**

- $K_a$ Karlovitz number
- $K_{a,t}$ turbulence Karlovitz number
- $L_e$ Lewis number
- $M$ Markstein number
- $M_a$ Mach number
- $Re_t$ turbulence Reynolds number
- $Sc$ Schmidt number
- $Ze$ Zeldovich number

**Miscellaneous**

- $[A_i]$ molar concentration of $A_i$ mol m$^{-3}$
- $\Delta t$ time step s
- $\Delta x$ mesh size m
Abstract

This thesis focuses on the numerical simulation of spherical and cylindrical premixed flames and on the investigation of various aspects of these flames, especially on flame movement and flame stretching. For the numerical simulation, two different approaches are used: a flamelet description and Direct Numerical Simulations (DNS).

In flamelet models, the fast chemical processes inside a flame are decoupled from the relatively slow movement of the entire flame itself by using a kinematic equation for the flame movement. In this thesis, such a flamelet model was developed to keep the computational costs relatively low while maintaining the computations accurate. Several parts of this flamelet model are different from existing ones: the flow field is obtained from the combination of the continuity equation and the kinematic equation for the density, the flame is considered to have a finite thickness instead of being infinitely thin, and the flame stretch rate takes the variation of the flame thickness into account. This flamelet model is suitable to simulate perfectly spherical and cylindrical flames that are weakly stretched, which means that these flames have a large radius compared to the flame thickness.

For the simulation of freely expanding flames in a turbulent flow field, a DNS program was developed. Such a program solves the equations that govern combustion directly. The main disadvantage of DNS is its very high computational cost, especially for large computational domains and complex chemistry. These costs were reduced by using both a small domain and a chemical reduction technique. Two different reduction techniques were used here: the single-step reaction mechanism and the Flamelet-Generated Manifold (FGM) method. The single-step reaction mechanism only contains the global reaction instead of a large number of elementary reactions, thereby reducing the number of differential equations that need to be solved. The FGM method computes multidimensional flames by using one-dimensional flame simulations.

These numerical flame simulations were used to investigate the behaviour of the spherical and cylindrical flames in more detail. It appeared that the burning velocity varies significantly through the flame, whereas the flame propagation velocity varies only slightly. This small variation is due to the decrease of the flame thickness during the expansion or implosion of the flames. Expansion causes the flame to endure a decreasing stretch, whereas implosion causes an increa-
ing compression. The parameter that takes these effects into account is the flame stretch rate, which is defined as the relative rate of change of an infinitely small volume in the flame. Besides the dependence on the flame movement, this flame stretch rate is also dependent on the position in the flame. The most appropriate position to evaluate the combustion variables appeared to be the inner layer, which is defined as the position in the flame where the most important combustion reactions take place. At this inner layer, the burning velocity of steady spherical and cylindrical flames was proven to be almost independent of the flame radius.

A very important aspect of the expanding and imploding flames is the influence of the stretching of these flames on the mass burning rate, which is the local laminar burning velocity multiplied with the local density. Therefore, detailed theoretical and numerical analyses of this relation were performed. Weakly stretched flames show a linear relation between the ratio of the stretched and unstretched mass burning rate and the flame stretch rate. The proportionality factor is the Markstein number, which is dependent on the position in the flame. So far, expressions for this number have been derived from asymptotic analyses. In this investigation however, expressions were derived from an integral analysis. These expressions give more accurate results than the asymptotic expressions, because the structure of the flame is taken into account. Furthermore, the Markstein numbers for curvature and flow straining were analysed. According to a phenomenological law, the Markstein number can be separated into a part for the curvature of the flame and a part for the straining of the flow. It was shown, however, that it is not possible to introduce a separate and unique Markstein number for the flow straining that can be used to describe its influence in different combustion situations.

The flamelet model and DNS program were shown to accurately simulate spherical and cylindrical flames that are weakly or strongly stretched. These simulations were successfully used to gather a more detailed insight in the behaviour of these flames.
Samenvatting

Dit proefschrift richt zich op de numerieke simulatie van bol- en cilindervormige voorgemengde vlammen en op het onderzoek naar verscheidene eigenschappen van deze vlammen, met name naar de beweging en oprekking ervan. Voor de numerieke simulatie zijn twee verschillende methoden gebruikt: een flamelet-beschrijving en Directe Numerieke Simulaties (DNS).

In flamelet-modellen worden de snelle chemische processen in een vlam ontkoppeld van de relatief langzame beweging van de gehele vlam door gebruik te maken van een kinematische vergelijking voor de beweging van de vlam. In dit proefschrift is zo’n flamelet-model ontwikkeld om ervoor te zorgen dat de rekentijd relatief kort blijft terwijl de resultaten toch nauwkeurig blijven. Meerdere delen van dit flamelet-model verschillen van bestaande flamelet-modellen: het snelheidsveld wordt berekend uit de combinatie van de continuïteitsvergelijking en de kinematische vergelijking voor de dichtheid, de vlam wordt verondersteld een eindige dikte te hebben in plaats van een infinitesimaal dunne, en in de uitdrukking voor de snelheid waarmee de vlam kan oprekken is de variatie van de vlamdikte opgenomen. Dit flamelet-model is geschikt om perfect bol- en cilindervormige vlammen te simuleren die zwak opgerekt zijn, hetgeen inhoudt dat deze vlammen een grote straal hebben ten opzichte van de vlamdikte.

Voor de simulatie van expanderende vlammen in een turbulent stromingsveld is een DNS-programma ontwikkeld. Zo’n programma lost de verbrandingsvergelijkingen direct op. Het belangrijkste nadeel van DNS is de zeer lange rekentijd, vooral als grote rekendomeinen en complexe chemie worden gebruikt. In dit onderzoek zijn de rekentijden verkort door het gebruik van een klein rekendomein en een chemische reductietechniek. Twee verschillende reductietechnieken zijn hier gebruikt: het eenstapsreactiemechanisme en de Flamelet-Generated Manifold (FGM) methode. Het eenstapsreactiemechanisme bevat alleen de globale reactie in plaats van een groot aantal elementaire reacties, waardoor het aantal differentiaalvergelijkingen dat opgelost moet worden drastisch is verminderd. De FGM-methode berekent meerdimensionale vlammen door gebruik te maken van eendimensionale vlamsimulaties.

Deze numerieke simulaties werden gebruikt om het gedrag van bol- en cilindervormige vlammen in meer detail te bestuderen. Het bleek dat de verbrandingsnstand door de vlam heen aanzienlijk varieert, terwijl de vlamsnelheid dit slechts beperkt doet. Deze kleine variatie is het gevolg van de afname van de
vlamdikte terwijl de vlam expandeert of implodeert. Expansie veroorzaakt een
afnemende oprekking van de vlam, terwijl implosie verantwoordelijk is voor een
toenemende samendrukking ervan. De parameter die deze effecten beschrijft is
de vlamreksnelheid, die is gedefinieerd als de relatieve verandering van een on-
eindig klein volume in de vlam. Naast de afhankelijkheid van de vlambeweging
is deze vlamreksnelheid ook afhankelijk van de positie in de vlam. De meest
geschikte plaats om de verbrandingsvariabelen te evalueren bleek de binnenste
reactielaag te zijn. Deze laag ligt op die plaats in de vlam waar de belangrijkste
verbrandingsreacties plaatsvinden. Het is bewezen dat de verbrandingsnauwkeurig
heid van stilstaande bol- en cilindervlammen bijna onafhankelijk is van de vlamstraal
van deze laag.

Een belangrijk aspect van expanderende en imploderende vlammen is de in-
vloed die de oprekking ervan heeft op de massaverbrandingssnelheid, die is ge-
definieerd als de lokale laminaire verbrandingssnelheid vermenigvuldigd met de
lokale dichtheid. Om die reden zijn er verschillende theoretische en numerieke
analyses naar deze relatie uitgevoerd. Zwak opgerekte vlammen vertonen een
lineaire relatie tussen de verhouding van de opgerekte en niet-opgerekte mas-
soeverbrandingsnauwkeurigheid en de vlamreksnelheid. De evenredigheidsfactor is het
Marksteingetal, dat afhankelijk is van de positie in de vlam. Tot dusver waren
uitdrukkingen voor dit getal steeds afgeleid van asymptotische analyses. In dit
onderzoek zijn echter uitdrukkingen afgeleid met behulp van een integraalanaly-
se, die tot nauwkeuriger resultaten leidden dan de asymptotische uitdrukkingen,
omdat er rekening gehouden is met de structuur van de vlam. Verder zijn de
Marksteingetingallen geanalyseerd die betrekking hebben op de kromming van de
vlam en de vervorming van het stromingsveld. Volgens een bestaande fenome-
nologische wet zou het Marksteingetal opgedeeld kunnen worden in een deel
dat verbonden is aan de kromming van de vlam en een deel dat een relatie heeft
met de vervorming van het stromingsveld. Het is hier echter aangetoond dat
het principieel niet mogelijk is om een uniek Marksteingetal voor de vervorming
van het stromingsveld te introduceren dat gebruikt kan worden om de invloed
op verschillende verbrandings situaties te beschrijven.

Het is aangetoond dat het flamelet-model en het DNS-programma bol- en ci-
lindervormige vlammen die zwak of sterk opgerekt zijn nauwkeurig kunnen si-
muleren. Deze simulaties werden met succes gebruikt om een meer gedetailleerd
inzicht in het gedrag van deze vlammen te verkrijgen.


Curriculum vitae

Sept. 10th, 1976 : Born in Eindhoven, the Netherlands.

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Sept. 1994 – June 1999 : Mechanical Engineering,
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