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Development of Flamelet Generated Manifolds for Partially-Premixed Flame Simulations

PROEFSCHRIFT

ter verkrijging van de graad van doctor aan de Technische Universiteit Eindhoven, op gezag van de rector magnificus, prof.dr.ir. C.J. van Duijn, voor een commissie aangewezen door het College voor Promoties in het openbaar te verdedigen op woensdag 19 oktober 2011 om 16.00 uur

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Nomenclature

A | Flame stretch transport equation | 135 |

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Acknowledgements
Abstract

Development of Flamelet Generated Manifolds for Partially-Premixed Flame Simulations

Accuracy of simulations of combustion processes does not only depend on a meticulous description of the turbulent flow field, its accuracy and detail depends on the representation of combustion chemistry and its interaction with turbulence as well. Simulations with a high level of exactness are very time consuming due to the large range in length- and timescales in turbulence and chemical kinetics. The contribution of solving the flow field to the computational cost can be reduced by only solving the major turbulent motions containing the largest part of the turbulent kinetic energy. In this method, the Large Eddy Simulation (LES) method, the influence of small, non-resolved eddies has to be modeled, but good models for this unresolved transport of momentum are readily available. Reducing the chemical kinetics is typically a more difficult job due to the large number of elementary (chemical) reactions to be taken into account and the associated stiffness of the system of equations due to the (very) large range in time scales. The Flamelet Generated Manifold (FGM) reduction method resolves this issue by the creation of a chemical manifold out of laminar one-dimensional flame structures. These one-dimensional flame structures, called flamelets, are computed using detailed chemistry and are subsequently tabulated in composition space as a function of a small number of control variables. During a numerical simulation of a flame, only transport equations for the control variables have to be solved. The FGM method can thus be interpreted as a combination of flamelet and manifold methods: the low-dimensional chemical manifold, which is used in simulations of multi-dimensional flames, is based on one-dimensional flame structures containing all transport and chemical phenomena as observed in three-dimensional flames.

A small number of archetypical flamelet types exist of which the premixed and counterflow diffusion are used most commonly. They represent premixed and non-premixed flames, two limiting combustion modes. Partially-premixed combustion is considered to occur somewhere between these two types and no archetypical flamelet type exists for stratified combustion. This doctoral dissertation focusses on the following main research question: can partially-premixed combustion be adequately modeled by FGM tables based on premixed flamelets, counterflow diffusion flamelets or a combination of both types? The importance of this research question lies hidden in the large number of industrially-relevant partially-premixed combustion applications for which the use of the FGM method in simulations can speed up design processes.

The use of the FGM reduction method can reduce computational costs with a few orders of magnitude [Oijen2002-2], but the requirement that the reaction layer should be resolved still claims a large number of grid points in the simulation of turbulent flames. The Flame
Surface Density (FSD) model allows much coarser grids to be used, but this method has been specifically defined for premixed flames. In chapter 3 it is shown that the FSD approach can also be used in partially-premixed flames having stratification levels as typically observed in gas turbines. This was investigated by performing Direct Numerical Simulations (DNS) of turbulent planar Bunsen flames, enabled by the use of the FGM method. A priori analysis of the DNS results indicate that simple Presumed PDF subfilter models for the filtered (surface-averaged) mass burning rate yield fairly accurate predictions when filter widths of up to eight flame thicknesses are used. The implementation of the FSD model using mass burning rate data from flamelets can therefore be considered to be a feasible possibility for the simulation of turbulent stratified flames in an industrial environment.

In chapter 4 the focus shifts from flames with only a moderate stratification towards flames with a larger range of equivalence ratios: the well-documented Sandia flames. For CO, CO$_2$, H$_2$, H$_2$O and OH mass fractions, a priori comparisons are made with experimentally obtained data from flames with Reynolds numbers ranging from 13,400 for the moderately turbulent flame C to 44,800 for flame F in which local quenching plays an important role. It is concluded that counterflow diffusion flamelet-based FGM’s prove to be significantly more accurate than premixed-based ones for H$_2$, CO$_2$ and CO mass fraction predictions. For fuel-rich conditions, premixed flamelet-based FGM’s tend to severely overestimate H$_2$ and CO mass fractions while underestimating CO$_2$ mass fractions. Preferential diffusion effects were only visible at low Reynolds numbers and close to the burner nozzle: in general the unit Lewis number assumption is an appropriate one for these flames.

Where all numerical and modeling errors in the Computational Fluid Dynamics (CFD) simulations have been excluded in the a priori analysis in chapter 4, in chapter 5 these are taken into account in LES of Sandia flame D and F. Simulation results appear to be very sensitive to boundary conditions for turbulent velocity fluctuations. However, when outcomes for the reaction progress variable, H$_2$ and CO are viewed in composition space, a fairly good resemblance with experimental results is obtained. Using additional transport equations for H$_2$ and CO instead of interpolating them directly from the FGM table does not improve results. A priori predictions of H$_2$ and CO do show a significant improvement in accuracy compared to LES results. It can be concluded that tabulated FGM chemistry can yield accurate predictions even for difficult species like H$_2$ and CO, provided that the CFD solver predicts control variable fields with high accuracy.

For all flames considered in chapters 3 and 5 the type of flamelets to use for the generation of the FGM table was clear, either from literature or from a priori analysis. Chapter 6 tackles the question whether FGM tables based on either premixed or non-premixed flamelets can be combined for the simulation of partially-premixed flames. In many industrially-relevant flames, it is simply not known beforehand which flamelet type is most applicable and therefore an adaptive method is highly desirable. Globally, the adaptive method does yield an improvement in flame structure predictions when it is unknown which flamelet structure, premixed or non-premixed, is closest to the partially-premixed flame structure. For H$_2$, the combination of two FGM tables by means of the proposed smooth transition function does improve results significantly. The fact that one FGM table overestimates $Y_{H_2}$ and the other underestimates this mass fraction contributes to the success of the adaptive method for this species. For CO both FGM tables overestimate $Y_{H_2}$, although using the premixed-flamelet based table, this overestimation...
substantially larger. The adaptive method yields values for $Y_{\text{H}_2}$ significantly closer to detailed chemistry results than the premixed flamelet-based FGM table does, but the non-premixed flamelet-based FGM obtains best results, especially for slightly rich conditions. Like in chapter 5, additional transport equations for $\text{H}_2$ and $\text{CO}$ in which the chemical source term comes from the combination of FGM tables does not improve results; direct interpolation from the FGM tables remains more accurate for these flames as well.

It can be concluded that the FGM method is a powerful tool which renders DNS and LES of complex flames possible. Without this reduction method, simulations discussed in chapter 3 and 5 would have taken amounts of computational resources which are usually not available. The FGM method obtains accurate predictions for species mass fractions, of which $\text{H}_2$ and $\text{CO}$ are mainly treated in this dissertation, provided that control variables are accurately predicted by the CFD solver and appropriate assumptions are made for the flamelets from which the FGM table is created. The latter however, requires a certain insight in combustion physics implying that using the FGM method in partially-premixed flames is not (yet) plug-and-play. The importance of an accurate CFD-solver together with meticulous boundary conditions for turbulent fluctuations is confirmed by the significant difference between a priori predictions and LES outcomes in chapter 5. Premixed flamelet-based and counterflow diffusion flamelet-based FGM tables can not be straightforwardly be combined for the description of partially-premixed one-dimensional flames. The method proposed in chapter 6 does improve results when no a priori knowledge is available, and flamelet modeling assumptions for a single FGM table would be taken randomly. For intermediate applied strain rates and equivalence ratio variation the combination of these two types of FGM tables consistently results in a better description of a partially-premixed flame. For other conditions the use of a single FGM table is generally obtains better predictions, provided that it is known which single FGM table should be used. If this knowledge is absent, the use of the combined FGM method is a safe method, in which errors in all cases discussed in this thesis are smaller than would be obtained using only the FGM table which obtained the most inaccurate predictions.
Samenvatting

Development of Flamelet Generated Manifolds for Partially-Premixed Flame Simulations

Nauwkeurigheid van simulaties van verbrandingsprocessen hangen niet alleen af van een nauwkeurige beschrijving van het turbulente stromingsveld, maar ook van een minutieuze beschrijving van de optredende chemische reacties en hun interactie met de turbulentie. Simulaties met een hoge nauwkeurigheid zijn erg kostbaar door de grote variatie in lengte- en tijdschalen in zowel de turbulente stroming als in de chemische reacties. De kosten die gemaakt moeten worden om het stromingsveld op te lossen kunnen gereduceerd worden door alleen de grote turbulente wervels op te lossen, die het voornaamste aandeel in de turbulente kinetische energie bezitten. In deze methode, Large Eddy Simulation (LES) genaamd, wordt de bijdrage van kleine, niet opgeloste turbulente wervels gemodelleerd; goede modellen hiervoor zijn in het verleden ontwikkeld. Het reduceren van het chemische reactieschema is moeilijker door het grote aantal elementaire chemische reacties die meegenomen moeten worden, en de hoge stijfheid van het stelsel van beschrijvende mathematische vergelijkingen door de grote variatie van de opgenomen tijdschalen. De Flamelet Generated Manifold (FGM) reductiemethode lost dit probleem op door gebruik te maken van getabelleerde chemie, gebaseerd op laminaire, eendimensionale vlamstructuren. Deze eendimensionale vlammen, genaamd flamelets, worden berekend met gebruik van gedetaileerde chemische reactieschema’s en worden vervolgens getableerd in compositie ruimte als een functie van een klein aantal controleanalogen. Tijdens de simulatie van een vlam hoeven bij gebruik van een FGM-tabel alleen transportvergelijkingen opgelost te worden voor deze controleanalogen. De FGM aanpak kan dus gezien worden als een combinatie van flamelet- en manifold-methoden: de laagdimensionele chemische manifold die gebruikt wordt in simulaties van twee- of driedimensionale vlammen, is gebaseerd op een-dimensionale vlamstructuren die zowel convectief als diffusief transport bevatten, alsmede een groot aantal chemische reacties zoals die optreden in werkelijke vlammen. Een klein aantal karakteristieke flamelet typen bestaat, waarvan de voorgemengde en niet-voorgemengde (counterflow) flamelets verreweg het meest bekend zijn. Zij representeren voorgemengde en niet-voorgemengde vlammen: de twee vlamtypen die in verbrandingsonderzoek van elkaar onderscheiden worden. Gedeeltelijk voorgemengde verbranding treedt op tussen deze twee limieten en er bestaat geen karakteristieke flameletstructuur voor dit type verbranding. Dit proefschrift richt zich op de volgende centrale onderzoeksvraag: kan gedeeltelijk voorgemengde verbranding nauwkeurig worden gemodelleerd met gebruik van FGM-tabellen die gebaseerd zijn op voorgemengde flamelets, niet-voorgemengde flamelets of een combinatie van deze twee? Het belang van deze onderzoeksvraag ligt in de grote hoeveelheid industriële toepassingen van gedeeltelijk voorgemengde vlammen waarvoor de FGM methode ontwikkel-
optimalisatieprocessen aanzienlijk zou kunnen versnellen.

Het gebruik van de FGM reductiemethode kan rekentijden met een paar ordes van grootte reduceren [Oijen2002-2], maar het vereiste dat de reactielaag opgelost word vergt nog steeds een groot aantal roosterpunten in simulaties van turbulente vlammen. Het Flame Surface Density (FSD) model maakt het mogelijk aanzienlijk grovere numerieke roosters te gebruiken, maar dit model is tot op heden specifiek ontwikkeld voor simulaties van voorgemengde vlammen. In hoofdstuk 3 wordt aangetoond dat de FSD aanpak ook voor gedeeltelijk voorgemengde vlammen toepasbaar is, met variaties in equivalentieverhouding die typisch in gasturbines voorkomen. In dit onderzoek worden uitkomsten van Direct Numerical Simulations (DNS) van turbulente Bunsen vlammen gebruikt, die uitvoerbaar werden door het gebruik van de FGM methode. *A priori* analyse van deze uitkomsten geven aan dat een eenvoudige Presumed PDF subfilter modellen voor de gefilterde (over het oppervlakte gemiddeld) consumptiesnelheid van de brandstof goede voorspellingen leveren als filterbreedtes tot acht vlamdiktes gebruikt worden. De implementatie van het FSD model, waarin brandstofconsumptiesnelheden uit flamelets gebruikt worden, kan daarom gezien worden als een veelbelovend model voor het gebruik in een ontwikkelings- en/of optimalisatieproces.

In hoofdstuk 4 verschuift de aandacht van vlammen met een geringe variatie in equivalentieverhouding naar vlammen waarin een grotere variatie optreedt: de goed gedocumenteerde Sandia vlammen. Voor CO, CO$_2$, H$_2$, H$_2$O en OH massafracties wordt een *a priori* vergelijk gemaakt tussen experimentele data en voorspellingen door FGM-tabellen in vlammen met (jet) Reynolds getallen tussen 13.400 en 44.800, waarbij in laatstgenoemde locale uitdoving en (her-)ontsteking een belangrijke rol speelt. FGM-tabellen gebaseerd op niet-voorgemengde flamelets blijken aanzienlijk betere voorspellingen voor H$_2$, CO$_2$ en CO massafracties te geven dan tabellen gebaseerd op voorgemengde flamelets. Onder brandstofrijke condities overschatten op voorgemengde flamelets gebaseerde FGM-tabellen H$_2$ en CO massafracties substantieel, terwijl waarden voor CO$_2$ onderschat worden. Effecten van preferentiële diffusie zijn alleen zichtbaar bij lage Reynolds getallen en in de buurt van de brander: voor deze vlammen is de aanname dat het Lewis getal gelijk is aan één een goede aannemer.

In de *a priori* analyse in hoofdstuk 4 zijn alle numerieke en modelleringsfouten uitgesloten, die in Computational Fluid Dynamics (CFD) simulaties wel gemaakt worden. In hoofdstuk 5 worden deze fouten meegenomen in LES van de Sandia vlammen D en F. Numerieke resultaten blijken erg gevoelig te zijn voor de manier waarop turbulente fluctuaties in de randvoorwaarden worden voorgeschreven. Desondanks, wanneer resultaten voor de voortgangsvariabele, H$_2$ en CO in compositieruimte bekeken worden, wordt er een betrekkelijk goede overeenkomst tussen experimentele data en numerieke uitkomsten gevonden. Indien er een extra transportvergelijking voor H$_2$ of CO gebruikt wordt, in tegenstelling tot het direct interpoleren van data uit de FGM-tabel, verbeteren resultaten niet. *A priori* voorspellingen voor Y$_{H_2}$ en Y$_{CO}$ zijn aanzienlijk beter dan LES resultaten, wat erop duidt dat FGM-tabellen nauwkeurige voorspellingen kunnen doen, zelfs voor moeilijke stoffen als H$_2$ en CO, onder voorwaarde dat de CFD-code de velden voor de controlevariabelen met een hoge nauwkeurigheid reproduceert.

Voor alle vlammen in hoofdstukken 3 en 5 was uit literatuur of uit *a priori* analyses duidelijk welk flamelet type het best gebruikt kon worden voor het maken van de
FGM-tabel. In hoofdstuk 6 wordt onderzocht of gedeeltelijk voorgemengde vlammen nauwkeurig gmodelleerd kunnen worden door combinatie van een FGM-tabel gebaseerd op voorgemengde flamelets en een tabel gebaseerd op niet-voorgemengde flamelets. In veel industriële toepassingen is het eenvoudigweg niet bekend welk flamelet type het best gebruikt kan worden en daarom is het wenselijk een adaptieve methode te gebruiken die locaal bepaalt met welke weegfactoren de verschillende tabellen gecombineerd moeten worden. De voorgestelde adaptieve methode levert nauwkeurigere voorspellingen van de voortgangsvariabele, indien het onbekend is welk flamelet type, voorgemengd of niet-voorgemengd, het best overeen komt met de gesimuleerde gedeeltelijk voorgemengde vlam. Voor H\textsubscript{2} verbeteren voorspellingen aanzienlijk als de voorgestelde combinatiemethode gebruikt wordt, in plaats van een enkele tabel gebaseerd op voorgemengde of niet-voorgemengde flamelets. Dit is voor een groot deel te danken aan het feit dat \(Y_{H_2}\) wordt overschat door de tabel gebaseerd op voorgemengde flamelets, en onderschat door de tabel gemaakt van niet-voorgemengde flamelet oplossingen. De combinatie van deze twee ligt dus altijd ergens in het midden, dichter bij de oplossing die gevonden wordt wanneer een gedetailleerd reactieschema gebruikt wordt. \(Y_{H_2}\) wordt door beide tabellen overschat, hoewel deze overschatting groter is voor de FGM-tabel gebaseerd op voorgemengde flamelets. De voorgestelde combinatiemethode voorspelt \(Y_{H_2}\)-waarden die veel dichter bij de gedetailleerde oplossing liggen dan wanneer alleen de op voorgemengde flamelets gebaseerde tabel gebruikt wordt, maar die onnauwkeuriger zijn dan wanneer alleen de op niet-voorgemengde flamelets gebaseerde tabel gebruikt wordt. De verschillen zijn voornamelijk zichtbaar onder brandstofrijke condities. Net zoals in hoofdstuk 5 is aangetoond, leveren transportvergelijkingen voor H\textsubscript{2} en CO waarin de chemische bronterm uit de FGM-tabellen gebruikt wordt, in plaats van de massafraction zelf, geen verbetering van de resultaten op: ook voor deze vlammen is directe interpolatie van massafracties uit FGM-tabellen de meest nauwkeurige methode.

Samenvattend kan worden gesteld dat de FGM methode een belangrijk stuk gereedschap is, dat DNS en LES van complexe vlammen mogelijk maakt. Zonder deze reductiemethode zouden de vlamsimulaties zoals besproken in hoofdstukken 3 en 5 praktisch onmogelijk geweest zijn. Met gebruik van de FGM methode kunnen nauwkeurige voorspellingen gedaan worden voor massafracties van bepaalde stoffen, waarvan met name H\textsubscript{2} en CO behandeld zijn in dit proefschrift. Dit is echter alleen mogelijk onder voorwaarde dat de CFD-code nauwkeurige voorspellingen voor de controlevariabelen doet, en dat er modelleringsaanname voor de gebruikte flamelets gedaan zijn die in overeenstemming zijn met het karakter van de te simuleren vlam. Dit laatste aspect vereist een minimaal inzicht in verbrandingsfysica, wat impliceert dat het gebruik van de FGM methode voor de simulatie van gedeeltelijk voorgemengde vlammen nog niet plug-and-play is. Het belang van het gebruik van een nauwkeurige CFD-code samen met nauwkeurig voorgeschreven randvoorwaarden voor turbulente fluctuaties, wordt bevestigd door de significante verschillen tussen a priori resultaten en LES uitkomsten in hoofdstuk 5. Tabellen gebaseerd op voorgemengde of niet-voorgemengde flamelets kunnen niet simpelweg gecombineerd worden voor de beschrijving van gedeeltelijk voorgemengde eendimensionale vlammen. De methode die voorgesteld wordt in hoofdstuk 6 verbetert voorspellingen indien er geen a priori kennis beschikbaar is, en modelleringsaannamen in de flamelets willekeurig gekozen worden. Voor gematigde opgelegde strain rates en gematigde niveaus van voormenging levert de gecombineerde FGM methode consistente verbetering op in de beschrijving van de voortgangsvariabele. Voor andere randvoorwaarden geeft het gebruik van een enkele FGM-tabel de nauwkeurigste
voorspellingen, indien de tabel gekozen wordt waarvan de flameletstructuur het beste overeenkomt met de te simuleren vlam. Indien deze a priori informatie niet beschikbaar is, is de gecombineerde FGM een veilige methode, waarin onnauwkeurigheden in alle in dit proefschrift besproken voorbeelden kleiner zijn dan onnauwkeurigheden die gevonden zouden worden indien de minst geschikte FGM-tabel gebruikt zou worden.
Introduction

1.1 Research background

Numerical simulations of flames have gained an increasing interest over the past few decades and simulation strategies have matured up to a point where they can be used to optimize engineering combustion appliances. The limited availability of (fossil) fuels and the formation of pollutants are driving forces for this optimization process. In industry, significant reductions in optimization time and associated expenses can be obtained by the replacement of experiments by simulations. Nevertheless, accurate predictions of mass burning rates, flame stabilization behavior and pollutant formation still pose a serious challenge for combustion engineers. This is due to the large number of species involved in chemical reactions and the multi-scale interaction between the flame and flow field which make accurate numerical simulations of reactive flow exceptionally complex.

In engineering combustion appliances, almost without exception turbulent flow behavior is observed. For accurate predictions of flow properties, these turbulent fluctuations need to be taken into account. For non-reactive flows this is already important, but for reactive flows this is even more significant since chemical kinetics react very non-linearly to small perturbations. To capture these turbulent fluctuations, Direct Numerical Simulation (DNS) and Large Eddy Simulation (LES) strategies were developed, but solving the (time-dependent) flow field using one of these methods requires substantial computing power. This is especially true for DNS where length scales from the integral length scale $L_{\text{INT}}$ down to the Kolmochorov length scale $\eta$ are resolved. For DNS of single-phase non-reactive flows it can be shown that the required number of grid points and increments in time are algebraically proportional to the integral Reynolds number $Re_{L_{\text{INT}}}$ [Pope2000]:

$$N_g \propto Re^{9/4}_{L_{\text{INT}}}, \quad N_t \propto Re^{1/2}_{L_{\text{INT}}},$$

implying that required computational resources are proportional to $Re^{11/4}_{L_{\text{INT}}}$. This requirement entails that DNS is impossible for nearly all engineering aerodynamic devices. This is where LES appears on the scene: in this approach only the major, kinetic energy containing, turbulent eddies are resolved, thereby reducing required computing power significantly. The influence of non-resolved turbulent eddies on the distribution of momentum, energy and species has to be modeled now, but LES enables the simulation of real-life aerodynamic appliances. For the development and validation of subfilter models for LES, DNS however remains an indispensable tool.

For simulations of reactive flow, additional challenges arise compared to non-reactive flow. The large number of involved species increases the number of transport equations to be solved by one or two orders of magnitude, while the large range in chemical time
Introduction

scales [Maas1992] complicate an accurate representation of the turbulence/chemistry interaction. The high stiffness and the large number of differential equations to be solved, make it difficult to obtain a numerical solution and methods to reduce the computing power requirements are highly desirable. Efficient reduction of reaction mechanisms, or a model to mimic reaction kinetics therefore has to be implemented. The use of detailed chemistry is preferred over the use of (empirical) models because of their generality and detailed information on pollutant formation, for example. The size of reaction mechanisms is generally proportional to the number of incorporated species and their accuracy in predicting species mass fractions, mass burning rates and ignition delays. For methane-air combustion, the GRI 3.0 mechanism [GRImech30] is one of the most comprehensive reaction mechanisms taking into account 53 species and 325 elementary reactions. The 57 transport equations that have to be solved (mass, momentum, energy and 52 species; the 53rd species is governed by conservation of mass) make this reaction mechanism very impractical or even impossible to use in DNS and LES. Smaller reaction mechanisms are available, but have their limitations in terms of accuracy and applicability. The trade-off between accuracy and feasibility of simulations has become apparent, and a sensible compromise will have to be met.

In combustion research, often premixed flames are distinguished from non-premixed flames. These two types exhibit a different character: in premixed flames chemical kinetics is rate-limiting while in non-premixed flames mixing, by means of diffusion or turbulence, can be considered the rate-limiting process. A large number of combustion models has been developed for application in LES and DNS; in many cases the model is specifically developed for either premixed or non-premixed flames. In many industrial combustors however, reactants are neither perfectly premixed, nor completely separated prior to combustion and this combustion mode is addressed as partially-premixed combustion. Chemical reaction mechanisms pose no explicit demands on their applicability to premixed or non-premixed combustion, but to properly capture chemical kinetics for both lean and rich conditions, many elementary reactions have to be taken into account. The application of smart reduction methods which preserve the detailed information on chemical kinetics of reaction mechanisms while reducing the required computing power significantly, are of key importance for the feasibility of LES in an industrial environment. The Flamelet Generated Manifold (FGM) reduction method theoretically can do this; in this thesis it is examined how it can be applied in the simulation of partially-premixed flames.

In this introductory chapter, first the governing equations that describe reactive flow are introduced. They will serve as a foundation for both the mathematical deduction of the FGM method and the DNS and LES simulations discussed in this thesis. An overview of reduction methods for chemical kinetics is given subsequently, pointing out the strengths and weaknesses of the FGM method compared to other reduction methods. At the end of this chapter, an outline of the succeeding chapters and their coherence is given.

1.2 Mathematical description of reacting flows

Flames can be seen as thin reaction-diffusion layers, embedded in a flow field. The governing equations of these reactive flows comprise convective, diffusive and (chemical) source contributions and their derivation can be found in many textbooks, for example [Williams1994, Warnatz1996, Poinsot2001].
1.2 Mathematical description of reacting flows

**General conservation equations**

Conservation of mass is expressed by the continuity equation:

\[
\frac{\partial \rho}{\partial t} + \frac{\partial \rho u_j}{\partial x_j} = 0 ,
\]

in which \( \rho \) denotes the mass density and \( u_j \) denotes velocity component \( j \) of the gas mixture. Conservation of momentum when only gravitation is considered as an external force is described by the Navier-Stokes equations:

\[
\frac{\partial \rho u_i}{\partial t} + \frac{\partial \rho u_i u_j}{\partial x_j} + \frac{\partial p}{\partial x_i} = \frac{\partial \tau_{ij}}{\partial x_j} + \rho g_i ,
\]

in which \( p \), \( \tau_{ij} \) and \( g_i \) denote the hydrostatic pressure, the viscous stress tensor and the gravitational acceleration, respectively. For Newtonian fluids the viscous stress tensor \( \tau \) is assumed to depend linearly on velocity gradients [White1991] while bulk viscosity, the viscous resistance against pure dilatation, is neglected [Warnatz1996]:

\[
\tau_{ij} = \mu \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) - \frac{2}{3} \mu \delta_{ij} \frac{\partial u_k}{\partial x_k} ,
\]

in which \( \mu \) denotes the mixture averaged viscosity, \( \delta_{ij} \) denotes the Kronecker delta.

Conservation of energy can be written in terms of either internal energy \( e \) or enthalpy \( h \). In this thesis \( h \) is used due to its convenience in the description of isobaric combustion where for a closed system thermodynamics dictates that \( \Delta h = 0 \). In the absence of an external heat source or sink, the transport equation for \( h \) reads:

\[
\frac{\partial \rho h}{\partial t} + \frac{\partial \rho h u_j}{\partial x_j} + \frac{\partial \rho Y_i U_{ij}}{\partial x_j} = \frac{\partial p}{\partial t} + u_j \frac{\partial p}{\partial x_j} + \frac{\partial q_j}{\partial x_j} + \tau_{kj} \frac{\partial u_k}{\partial x_j} ,
\]

in which \( q_j \) denotes the diffusive energy flux in direction \( j \). In combustion applications the viscous dissipation term, i.e. the last term in equation (1.5), is often neglected due to its minor contribution. For chemically reacting flows, in addition to the equations (1.2-1.3 and 1.5) describing non-reacting flows, the non-constant composition of the gas mixture has to be described. This introduces additional transport equations for species mass fractions:

\[
\frac{\partial \rho Y_i}{\partial t} + \frac{\partial \rho Y_i u_j}{\partial x_j} + \frac{\partial \rho Y_i U_{ij}}{\partial x_j} = \dot{\omega}_i , \quad i \in [1, N_s] ,
\]

in which \( Y_i \) and \( \dot{\omega}_i \) denote the species mass fraction for species \( i \) and the chemical source term for species \( i \), respectively. Chemical reactions can not alter the total amount of mass, which results in the constraint

\[
\sum_{i=1}^{N_s} \dot{\omega}_i = 0 .
\]

\( U_{ij} \) denotes the diffusive velocity of species \( i \) in direction \( j \). Species mass fractions are defined as \( Y_i = \rho_i / \rho \) with \( \rho_i \) the density of species \( i \). Since chemical reactions can neither produce nor consume mass, the sum of chemical species source must be equal to zero. For reacting flows the diffusive energy flux \( q_j \) in equation (1.5) contains a contribution due to heat diffusion and a contribution associated with species diffusion [Williams1994]:

\[
q_j = -\lambda \frac{\partial T}{\partial x_j} + \rho \sum_{i=1}^{N_s} (h_i Y_i) U_{ij} ,
\]
in which $\lambda$ denotes the thermal conduction coefficient, which depends on the local temperature and composition, and $T$ denotes absolute temperature. The system of equations (1.2-1.3 and 1.5-1.6) is closed by two equations of state: the caloric equation of state and the thermal equation of state. The caloric equation of state, 

$$h = \sum_{i=1}^{N_s} Y_i h_i = \sum_{i=1}^{N_s} Y_i \left[ h_i^{\text{ref}} + \int_{T_{\text{ref}}}^{T} c_{p,i} \left( T' \right) \, dT' \right],$$  

(1.9)

defines the total enthalpy $h$ as a function of gas mixture composition, expressed by species mass fractions $Y_i$, and temperature $T$. The specific heat capacity $c_{p,i}$ of individual species is well documented in polynomial form [Kee1986]. Taking the gradient of $h$ in equation (1.9) and substitution of the result in equation (1.8) yields the following expression for the heat flux $q_j$:

$$q_j = -\frac{\lambda}{c_p} \frac{\partial h}{\partial x_j} + \sum_{i=1}^{N_s} h_i \left( \rho Y_i U_{ij} + \frac{\lambda}{c_p} \frac{\partial Y_i}{\partial x_j} \right), \quad \text{with} \quad c_p = \sum_{i=1}^{N_s} Y_i c_{p,i} \left( T \right),$$  

(1.10)

in which $c_p$ denotes the weighted specific heat at constant pressure. Gas mixtures are assumed to behave as an ideal gas and the thermal equation of state is then given by the ideal gas law:

$$\rho = \frac{pM}{(RT)} , \quad \text{with} \quad \bar{M} = \left( \sum_{i=1}^{N_s} Y_i \frac{M_i}{M} \right)^{-1},$$  

(1.11)

in which $R$ denotes the universal gas constant and $\bar{M}$ denotes the mean molecular mass.

**Diffusion of species**

Diffusion of species is governed by concentration gradients, pressure gradients and the influence of external forces [Williams1958]. For isobaric combustion without external forces, diffusion velocities of species can be computed by solving the system of equations [Ern1994]:

$$\frac{\partial X_i}{\partial x_k} = \sum_{j=1}^{N_s} X_i X_j \frac{D_{ij}}{D_{ij}} (U_{jk} - U_{ik}) + \sum_{j=1}^{N_s} \left[ \left( \frac{X_i X_j}{\rho D_{ij}} \right) \left( \frac{D_j}{Y_j} - \frac{D_i}{Y_i} \right) \right] \frac{\partial \log T}{\partial x_k},$$  

(1.12)

in which $D_{ij}$ represents the binary diffusion coefficients [Hirschfelder1954, Bird1960] and the last term in the right-hand side of the equation represents species diffusion due to temperature gradients, also known as the Soret effect. When the Soret effect is neglected, which is common practice for atmospheric combustion of hydrocarbons [Williams1994], equation (1.12) reduces to the well-known Stefan-Maxwell equation [Monchick1966]. The evaluation of equation (1.12) is very CPU-intensive and is therefore often replaced by a Fickian expression [Warnatz1996] for diffusive fluxes:

$$\rho U_{i,j} Y_i = -\rho D_i \frac{\partial Y_i}{\partial x_j}, \quad i \in [1, N_s],$$  

(1.13)

in which $D_i$ denotes the mixture-averaged diffusion coefficient. The introduction of the Lewis number

$$Le_i (x_j, t) = \frac{\lambda}{c_p (\rho D_i)^{-1}}, \quad i \in [1, N_s],$$  

(1.14)
which formally is a function of spatial coordinate $x_j$ and time $t$, allows the simplification of the species conservation equation (1.6) to:

$$\frac{\partial \rho Y_i}{\partial t} + \frac{\partial \rho Y_i u_j}{\partial x_j} = \frac{\partial}{\partial x_j} \left[ \frac{\lambda Y_i}{L_e c_p} \frac{\partial Y_i}{\partial x_j} \right] + \dot{\omega}_i, \quad i \in [1,N_s],$$

(1.15)

and for the heat flux $q_j$ can now be written:

$$q_j = -\frac{\lambda}{c_p} \left[ \frac{\partial h}{\partial x_j} + \sum_{i=1}^{N_s} \left( \frac{1}{L_e} - 1 \right) h_i \frac{\partial Y_i}{\partial x_j} \right].$$

(1.16)

From equation (1.16) it can be seen that if Lewis numbers for all species equal unity, the diffusive heat flux is only caused by a gradient of enthalpy.

When equation (1.6) is summed over all species, equation (1.2) is subtracted and constraint (1.7) is taken into account, the following restriction is obtained:

$$\sum_{i=1}^{N_s} \left( \frac{\lambda Y_i}{L_e c_p} \right) \frac{\partial Y_i}{\partial x_j} = 0.$$  

(1.17)

This implies that the $L_e$ for the abundant species does not have to be computed, but should be obtained using constraint (1.17) for consistency. In the remainder of this thesis, this constraint will not be specifically mentioned, but it is employed in computations.

### Combustion chemistry

In species conservation equation (1.15), the nett source term $\dot{\omega}_i$ accounts for production or consumption of species $i$ by chemical reactions. To illustrate how $\dot{\omega}_i$ is composed, the nett source term for carbon monoxide (CO) is computed using the two-step reaction mechanism for methane (CH₄) combustion proposed by Westbrook and Dryer [Dryer1972, Westbrook1981]. This two-step mechanism contains a non-reversible elementary reaction in which CO is formed as an intermediate species and a second reversible elementary reaction in which CO is oxidized to carbon di-oxide (CO₂):

$$\text{CH}_4 + 1.5 \text{ O}_2 \xrightarrow{k_1} \text{ CO} + 2 \text{ H}_2\text{O} ,$$  

(1.18)

$$\text{CO} + 0.5 \text{ O}_2 \xrightarrow{K_2} \xrightarrow{k_3} \text{ CO}_2 .$$  

(1.19)

In a homogenous system, for the nett chemical source term for CO the following expression is obtained:

$$\frac{\dot{\omega}_{CO}}{M_{CO}} = \left[ k_1 \left( \frac{\rho Y_{CH_4}}{M_{CH_4}} \right) \left( \frac{\rho Y_{O_2}}{M_{O_2}} \right)^{1.5} - k_2 \left( \frac{\rho Y_{CO}}{M_{CO}} \right) \left( \frac{\rho Y_{O_2}}{M_{O_2}} \right)^{0.5} + k_3 \left( \frac{\rho Y_{CO_2}}{M_{CO_2}} \right) \right].$$

(1.20)

The reaction rate coefficient $k$ is usually defined by a modified Arrhenius expression [Warnatz1996]:

$$k_j = A_j T^{\beta_j} \exp \left( -E_{A,j} / RT \right), \quad j \in [1,N_i],$$

(1.21)

where $N_i$ denotes the number of elementary reactions. For a reversible reaction like (1.19), the reverse reaction rate constant is related to the forward rate constant through the equilibrium constant $k_{eq}^j = k_j^f / k_j^r$ to ensure that the correct chemical equilibrium
composition is achieved. The equilibrium constant is well defined by thermodynamic properties of the species which are involved in the chemical reaction in tabulated form [Stull1971] or in polynomial form [Gordon1971, Kee1986]. A generic reversible chemical reaction can be written as:

\[
\sum_{i=1}^{N_s} \nu_{ij} M_i \rightleftharpoons \sum_{k=1}^{N_s} \nu''_{ij} M_k, \quad j \in [1, N_r]
\]  

(1.22)

in which \(\nu_{ij}\) and \(\nu''_{ij}\) are forward and backward stoichiometric coefficients for species \(i\) in reaction \(j\) and \(M_i\) represents species \(i\). The nett source term \(\dot{\omega}_i\) of species \(i\) for generic reaction (1.22) is composed of the contributions of each elementary chemical reaction:

\[
\dot{\omega}_i = M_i \sum_{j=1}^{N_r} \left( \nu''_{ij} - \nu_{ij} \right) r_j, \quad i \in [1, N_s],
\]  

(1.23)

in which \(r_j\) is the reaction rate of reaction \(j\). Since total mass must be conserved, the constraint that the sum of \(\dot{\omega}_i\) must equal zero, is obtained. The reaction rate of reaction \(j\) is expressed by:

\[
r_j = k_j \prod_{i=1}^{N_s} \left( \frac{\rho Y_i}{M_i} \right)^{\nu_{ij}} - k''_j \prod_{i=1}^{N_s} \left( \frac{\rho Y_i}{M_i} \right)^{\nu''_{ij}}, \quad j \in [1, N_r],
\]  

(1.24)

Chemical reactions can not produce or consume elements, they merely recombine elements into other molecules than were present in the initial state. For definitions which will be introduced in chapter 2, it is convenient to introduce the definition of an element mass fraction:

\[
Z_k = \sum_{i=1}^{N_s} \left( \frac{a_{ki} M_k}{M_i} \right) Y_i, \quad k \in [1, N_e],
\]  

(1.25)

in which \(a_{ki}\) denotes the number of atoms \(k\) in species \(i\), \(M_k\) denotes the molar mass of element \(k\) and \(M_i\) denotes the molar mass of species \(i\).

**Combustion equations**

The final set of partial differential equations describing reacting flow, in which viscous dissipation and external heat sources and sinks are neglected, can be recast as:

\[
\frac{\partial p}{\partial t} + \frac{\partial \rho u_i}{\partial x_j} = 0, \quad \text{(1.26)}
\]

\[
\frac{\partial \rho u_i}{\partial t} + \frac{\partial \rho u_i u_j}{\partial x_j} = -\frac{\partial p}{\partial x_i} + \frac{\partial \tau_{ij}}{\partial x_j} + \rho g_i, \quad \text{(1.27)}
\]

\[
\frac{\partial \rho Y_i}{\partial t} + \frac{\partial \rho Y_i u_j}{\partial x_j} = -\frac{\partial}{\partial x_j} \left[ \lambda \frac{\partial Y_i}{Le_i c_p \partial x_j} \right] + \dot{\omega}_i, \quad i \in [1, N_s],
\]  

(1.28)

\[
\frac{\partial \rho h}{\partial t} + \frac{\partial \rho h u_j}{\partial x_j} = \frac{\partial p}{\partial t} + \frac{\partial}{\partial x_j} \left[ \lambda \frac{\partial h}{c_p \partial x_j} + \frac{\lambda}{c_p} \sum_{i=1}^{N_s} \left( \frac{1}{Le_i} - 1 \right) h_i \frac{\partial Y_i}{\partial x_j} \right].
\]  

(1.29)

This renders the set of equations to a system of \(N_s + 5\) transport equations in which the highly non-linear chemical source term \(\dot{\omega}_i\) causes a strong coupling. The combination of the conservation of mass, equation (1.26), and conservation of each species, equation (1.28), over-determines the set of equations. To overcome this issue, for the abundant species, which in the case of hydrocarbon-air combustion is \(N_2\), \(Y_i\) is computed by the constraint:
reducing the system to \(N_s + 4\) transport equations. The number of species is generally quite large: for methane-air combustion, the number of species included in reaction mechanisms is of \(O(10^3)\) to \(O(10^2)\). The large range in chemical time scales for different species makes the set of equations (1.26-1.28) a very large, stiff system of equations.

1.3 Reduction methods for combustion chemistry

As pointed out in the previous section, to perform flame simulations conservation equations for mass, momentum, energy and the species in the reaction mechanism have to be solved simultaneously. The number of species is generally quite large and the highly non-linear chemical source term causes a strong coupling between different transport equations. The large range in chemical time scales [Maas1992] causes the governing set of equations to be stiff, requiring very small increments in time when an explicit solver is used. The introduction of large parallel computing approaches enables the use of detailed chemistry in simulations of real-life flames [Sankaran2007, Thevenin1996] while keeping computational wall-clock time to an acceptable level. In specific cases local grid refinement methods [Bell2007, Day2003] have been used to increase grid resolution only in regions where needed keeping the numerical grid coarse in regions of less interest. Multi-grid methods [Vreman2008] have also been implemented to speed up computations by using coarse-grid solutions as predictor steps for finer numerical grids.

Another approach to reduce computational requirements of flame simulations with detailed chemistry is to simplify the chemical reaction model. Steady-state species can be removed to reduce the number of required transport equations and to decouple the smallest chemical time scales, which relieves the restrictions on the timestep in explicit solvers. Reaction mechanism (1.18-1.19) can be interpreted as a reduced version of the previously introduced GRI 3.0 mechanism. Examples of “intermediately” reduced versions of the GRI 3.0 mechanism comprise the GRI 1.2 mechanism [GRImech12] (31 reactive species, 175 elementary reactions), the DRM19 mechanism [DRM19] (19 reactive species, 84 elementary reactions) and the ARM2 mechanism [Sung1998] (19 reactive species, 15 elementary reactions). The applicability of these reduced mechanisms is mostly limited to a specific range of conditions they are developed for. Outside this specific range, unrealistic results may be obtained since the relevant combustion chemistry is no longer included or reaction coefficients are tuned on results within the intended range of applicability.

When information on detailed reaction kinetics for a large range of conditions is required, for example in pollutant formation processes, mechanism reduction by removing species or elementary reactions is not an option. The observation that many chemical time scales are significantly smaller than flow time scales [Maas1992], allows decoupling of these fast chemical reactions. The ratio between the characteristic flow time scale \(\tau_f\) and chemical time scale \(\tau_c\) is conveniently expressed by the Dahmkohler number: \(Da = \tau_f / \tau_c\). A Quasi-Steady State (QSS) assumption [Peters1991] for species \(i\), i.e. \(\dot{\omega}_i = 0\), originates from homogeneous reacting system characterization, but is also applicable in flames for species with (very) high production and consumption rates, i.e. \(Da \gg 1\). Convective and diffusive time scales are then much larger than time scales associated with productive and
consumptive chemical time scales. These species can be considered slaved by slow-evolving species: they adapt very fast to changes in mass fractions of slow-evolving species. As a consequence, these steady-state species can be removed from the reaction mechanism reducing both the size and stiffness of the set of equations to be solved. It must be noted that solving the set of non-linear algebraic equations for the QSS species causes a small offset in the computational gain. The difference in chemical time scales is illustrated for a steady, unstrained premixed flame. In this case the convective flux, diffusive flux and chemical source of species \( i \) should balance according to:

\[
\frac{\partial \rho Y_i u_j}{\partial x_j} - \frac{\partial}{\partial x_j} \left[ \frac{\lambda}{Le_i c_p} \frac{\partial Y_i}{\partial x_j} \right] = \dot{\omega}_i = \dot{\omega}_i^+ + \dot{\omega}_i^- , \quad i \in [1, N_s] ,
\]

in which \( \dot{\omega}_i \) is the nett production rate which can be split into two parts: one containing all productive contributions, \( \dot{\omega}_i^+ \), and one containing all consumptive contributions, \( \dot{\omega}_i^- \). Figure 1.1 shows these fluxes for nitrogen monoxide (NO) and the hydroxyl (OH) radical in a stoichiometric premixed CH\(_4\)-air flame. Ambient conditions (\( T_0 = 300 \) K, \( p = 101.325 \) kPa) have been used and reaction kinetics are computed using the GRI 3.0 reaction mechanism [GRImech30] with unit Lewis numbers for all species. From figure 1.2 it can be seen that for NO the nett production rate is comparable to either the production or (absolute) consumption rate. For OH the production or (absolute) consumption rate are significantly larger than the nett production rate. It is concluded that for OH a QSS is appropriate, while it is not for NO.

For adiabatic flames, the accessible composition space is \( (N_s - N_e + 1) \)-dimensional: conservation of element mass fractions \( Z_k \) reduces dimensionality while pressure fluctuations add one dimension. The observation that a small number of rate-limiting reactions dictate the overall reaction progress results in the manifold concept: an attracting chemical manifold is assumed to exist, which can be parameterized by a number of variables \( N_v < (N_s - N_e + 1) \). For these slow evolving variables a transport equation has to be solved, the other variables can subsequently be retrieved from the manifold. In this class of methods, the reduction method proposed by Peters and Williams [Peters1987-2, Peters1991], the Intrinsic Low Dimensional Manifold (ILDM) method [Maas1992] and the Computational Singular Perturbation (CSP) method [Goussis1994] are the best-known mathematically-based methods. The method by Peters and Williams does not identify the QSS species or elementary reactions in Partial Equilibrium (PE) automatically, thereby requiring a thorough insight in reaction kinetics by the user. Both ILDM and CSP perform a time scale analysis, based on the local Jacobian of the chemical source term, automatically. The number of species which can be assumed in QSS typically varies along the trajectory in composition space. All three methods have in common that they only take into account the homogeneous set of equations containing the chemical source term, \textit{i.e.} advection and diffusion are fully neglected. When diffusive processes become increasingly important, \textit{e.g.} in cooler regions or during extinction and re-ignition processes, the dimensionality of the chemical manifolds rapidly increases and poor species mass fraction predictions are obtained [Oijen2000]. Time scales associated with production and consumption of slow evolving species like CO can be of the same order of magnitude as time scales associated with convective and diffusive processes [Maas1992, Poinson2001], resulting in poor predictive capabilities of these reduction techniques.

A different type of reduction techniques is formed by those methods which assume that trajectories in composition space in multi-dimensional flames closely resemble trajectories
1.3 Reduction methods for combustion chemistry

**Figure 1.1:** Comparison of fluxes for NO (left) and OH (right) in a steady one-dimensional premixed, stoichiometric methane/air flame at ambient initial conditions. Solid lines without markers indicate convective fluxes (\(\nabla \cdot (\rho \mathbf{u} Y_i)\)), solid lines with open markers indicate diffusive fluxes (\(\nabla \cdot \left( \frac{\lambda}{L_\epsilon c_p} \nabla Y_i \right)\)) and solid lines with solid markers indicate nett chemical sources (\(= \dot{\omega}_i\)).

**Figure 1.2:** Comparison of chemical sources for NO (left) and OH (right) in a steady one-dimensional premixed, stoichiometric methane/air flame at ambient initial conditions. Solid lines without markers indicate nett chemical sources (\(= \dot{\omega}_i\)), solid lines with open markers indicate total consumption rates (\(= \dot{\omega}_i^-\)) and solid lines with solid markers indicate total production rates (\(= \dot{\omega}_i^+\)).

of laminar, one-dimensional flames, denoted by the name flamelet [Peters1984]. This assumption holds when quasi one-dimensional flame structures can be isolated in multi-dimensional flames, *i.e.* when a main direction can be identified in which gradients of thermochemical variables are a few orders of magnitude larger than in directions perpendicular to this main one. The multi-dimensional flame can then be interpreted as an ensemble of flamelets embedded in a flow field. Even for turbulent flames this assumption remains valid when turbulent eddies cannot intrude the reaction layer: *e.g.* when flames in the corrugated flamelet regime and the thin reaction zones regimes [Borghi1988] are considered. The Flamelet Generated Manifold (FGM) [Oijen2000], or Flamelet Prolongated ILDM (FPI) [Gicquel2000], reduction methods, which are basically identical, identify a low-dimensional manifold based on flamelet structures and can therefore be considered as a combination of classic flamelet- and manifold methods. Extensions have been made to the ILDM method to include diffusive transport, resulting in the Phase-Space ILDM (PS-ILDM) [Bongers2002] and Reaction Diffusion Manifold (REDIM) [Bykov2006, Bykov2007] method for example. The FGM reduction method generates a database of thermochemical variables for specific initial conditions. Variables are stored as a function of a small number of controlling variables; in the ILDM method these are the slowest evolving, and thereby rate-determining species. During the flame simulation, only transport equations have to be solved for the controlling variables and dependent thermochemical variables can be retrieved from the stored chemical manifold.
Besides structured tabulation, which is common practice within the FGM method, data storage methods like Artificial Neural Networks [Blasco1999, Christo1996] and high-order orthogonal polynomial parameterizations [Turanyi1994] have been proposed to reduce required computer memory. These methods do not exclude one another and may be used in a combined approach. For the moment, the way the database is stored is not specified; in section 2.3.2 available methods will be compared briefly to point out their strengths and weaknesses.

Up-to-date flamelet-based reduction methods have been applied, in both laminar and turbulent flame simulations, with great success. In laminar flames, FGM tables composed of steady premixed flamelets have proven to be very accurate for Bunsen-type flames including heat loss effects [Oijen2000], highly stretched premixed counterflow flames [Oijen2002-1] and confined triple flames [Oijen2004]. In DNS of an expanding turbulent flame using the FGM method to represent combustion chemistry [Oijen2005], the use of a single control variable gave accurate predictions of the local mass burning rate. In LES of the well-known quasi non-premixed, turbulent Sandia Flames [Barlow2003], FGM tables composed of counterflow diffusion flamelets [Pitsch2000, Vreman2008] obtained significantly better species mass fraction predictions than could be obtained by FGM tables composed of steady premixed flamelets [Bradley1988, Vervisch2004]. This points out exactly one of the major issues in the application of flamelet methods in simulations of partially-premixed flames: an a priori choice has to be made for the flamelet type which is used to create the database. Premixed and counterflow diffusion flamelets are the two most distinct types of flamelets: they can be interpreted to represent two limits of a broad spectrum of partially-premixed flamelets [Talley1992].

In order to distinguish between premixed and non-premixed combustion, Yamashita et al. [Yamashita1996] introduced the flame-index $\xi$ based on the alignment of the spatial gradient of both fuel and oxidizer. Fiorina et al. [Fiorina2005-1] showed that the definition by Yamashita et al. fails in specific partially-premixed counterflow flames in which spatial gradient of both fuel and oxidizer are aligned, indicating premixed combustion, but combustion is still diffusion-controlled, indicating a non-premixed behavior. A modified version of $\xi$ is proposed but the new definition still has the setback that it has a binary behavior: no smooth transition between premixed and non-premixed combustion can be identified. Knudsen and Pitsch [Knudsen2009] proposed a method that distinguishes premixed from non-premixed combustion by considering how chemical source terms are balanced by diffusive and time-dependent terms. This method obtained good results in a posteriori analysis of a laminar triple flame and when applied in an LES of a premixed swirl burner.

However, the extend to what fuel and oxidizer are homogenously mixed prior to combustion in real-life combustors is often not known beforehand, e.g. for many gas turbine combustors or industrial furnaces no data is available a priori to support the choice of the most applicable flamelet type. In these applications, the reactants are neither perfectly premixed, nor completely non-premixed prior to combustion. This implies that the FGM user has to make this decision which can influence simulation outcomes, especially in terms of species mass fractions. The main issue which is addressed in this thesis, is the question which type of flamelet is most appropriate to model partially-premixed flames. Does a combination of those types yield a better description for partially-premixed flames?
1.4 Purpose of this study

The objective of this thesis is to assess the accuracy of low-dimensional FGM tables for turbulent, partially-premixed flames. The FGM method is originally based on premixed flamelets, but non-premixed flamelets can just as well be used to construct low-dimensional tables. Unfortunately for many, especially industry-relevant flames, no data is available to support the a priori choice which flamelet to use for the generation of the FGM database. On the other hand, species mass fractions, like $Y_{CO}$, strongly depend on the type of flamelet, see for example [Vreman2008]. Thereby the choice of the FGM user which type of flamelet to use, unintentionally influences the simulations outcome. This dissertation focusses on the question how to determine the most appropriate flamelet type to generate the FGM table from, and subsequently on the use of this table in DNS or LES of turbulent flames. An effort will be made to introduce an adaptive method, which determines the weight factors of two different FGM tables based on local conditions.

First, the FGM method is applied to simulations of flames with only small variations in equivalence ratio $\phi$. For these flames, previous studies have shown that premixed flamelets can accurately predict thermochemical variables [Oijen2004]. From an industrial point of view, it will be examined whether computational time can be further reduced by the use of the Flame Surface Density (FSD) model, which does suffer from the restriction that the reactive layer has to be resolved, like in the FGM approach.

Subsequently the range of $\phi$ present in the flame is increased and it is investigated whether an optimal flamelet type for FGM generation can be appointed when a priori knowledge, i.e. experimentally obtained data, is available. This a priori analysis is performed for the well-known Sandia Flames [Barlow2003] which behave as non-premixed flames but for which it is hypothesized that premixing plays an important role when flow-straining, quenching and re-ignition become more dominant [Bradley2002]. This hypothesis implies that in some regions a counterflow diffusion flamelet-based FGM is most applicable while in other regions a premixed flamelet-based FGM yield the best representation of detailed combustion chemistry. It is investigated whether such a bi-modal combustion behavior can be observed which would require the combination of different FGM tables in order to obtain an accurate description of the flame throughout the entire domain.

The next step is the actual use of the most applicable FGM table for the simulation the turbulent, partially-premixed Sandia Flames D and F. Since the thermochemical variables of main interest in this thesis are $Y_{CO}$ and $Y_{H_2}$, the table which obtains best predictions for these parameters in the a priori analysis is designated "most applicable" here. For these flames, DNS is practically impossible due to their high Reynolds numbers. LES will be used instead, and subfilter contributions have to be modeled. Compared to the a priori analysis, transport equations are now solved for the controlling variables giving rise to additional inaccuracies. Predictions of CO and $H_2$ mass fractions in these flames are generally not very satisfactory: it is assessed whether these mass fractions should be interpolated directly from the FGM table or whether using its source term in an additional transport equation yields improvement of the mass fraction predictions.

To overcome the difficulty of not knowing which type of FGM table is most applicable beforehand, it is examined whether the combination of counterflow diffusion flamelet-based and premixed flamelet-based FGM tables can accurately represent combustion chemistry in one-dimensional, partially-premixed flames. This is the final step in this research and for this purpose partially-premixed flamelets with different ranges in $\phi$ are computed using detailed chemistry and a combined FGM method. If results would
have shown an unambiguous and consistent improvement in one-dimensional flames, extension to three-dimensional flames would have been a recommendation for future research.

The main goal of the FGM reduction method is to reduce the computational cost of flame simulations without sacrificing too much accuracy. The majority of simulations presented in this thesis would not have been possible without the use of a reduction method for combustion chemistry. It is assessed whether the specific FGM reduction technique can successfully be used for partially-premixed combustion as well as premixed combustion, for which it has already proven its applicability.

1.5 Thesis outline

In previous sections the mathematical description of chemically reacting flows has been presented, and computational issues concerning this large, highly coupled set of equations have been pointed out. A brief introduction of reduction methods for combustion chemistry has been given, pointing out that all methods have their strengths and weaknesses. The FGM reduction method has already proven itself for being able to reduce required computational resources for premixed flames; the question tackled here is whether it can be adapted to yield accurate predictions for partially-premixed combustion as well.

Chapter 2 explains the FGM method: first the system of (generic) flamelet equations is derived from the combustion equations (1.26-1.28). In reality, the generic flamelet equations are recast in specific, premixed or non-premixed, forms before solving them; these specific equations and their boundary conditions are discussed in section 2.2. The parametrization, construction and utilization of FGM tables is finally explained in chapter 2. The applicability of premixed flamelet-based FGM tables for the simulation of stratified, i.e. partially-premixed flames with only a moderate variation in $\phi$, is examined in chapter 3. It is also investigated whether the Flame Surface Density (FSD) model for LES, originally intended specifically for premixed combustion, can be extended for partially-premixed combustion as well.

Chapter 4 shifts to flames having a higher variation in $\phi$: predictions from FGM tables generated using different modeling assumptions are analyzed for the well-known Sandia Flames C, D, E and F [Barlow2003] in an a priori analysis. Predictions for $\text{H}_2$, $\text{H}_2 \text{O}$, $\text{CO}$, $\text{CO}_2$ and OH mass fractions from premixed or non-premixed flamelet-based FGM tables are compared with measurements and it is examined whether the influence of preferential diffusion should be included in the FGM tables.

Chapter 5 deals with the question whether for CO and $\text{H}_2$ mass fractions, which are known to be hard to estimate accurately in rich conditions [Dally2003], predictions can be improved for real-life partially-premixed flames. In high-fidelity LES of Sandia Flames D and F predictions from solving an additional transport equation for these species will be compared with direct interpolation from FGM tables of these species.

In chapter 6 it is investigated whether combustion chemistry in one-dimensional partially-premixed flames can be approximated by a combination of premixed and non-premixed flamelet-based FGM tables. Comparisons will not only be made for the flame structure but also for $\text{H}_2$, CO, $\text{CO}_2$ and OH mass fractions.

Finally, summarizing conclusions are given in chapter 7.
FGM reduction method

This chapter treats the flamelet theory on which the Flamelet Generated Manifold reduction method is based. After deducing the flamelet equations, it is explained how these flamelet solutions can be combined into a manifold and how this manifold can subsequently be used in simulations of multi-dimensional flames.

Although Flamelet Generated Manifolds (FGM) were originally based on steady, adiabatic, freely-propagating premixed flamelets [Oijen2002-2] efforts have been made to unify the premixed and diffusion flamelet theory [Delhaye2009]. In many practical applications, the reactants are neither perfectly premixed, nor completely separated prior to combustion. Premixed and non-premixed flames can thus be interpreted to represent the two limits of a broad spectrum of flame structures [Talley1992]. For optimal use in (turbulent) flame simulations, the flamelet type on which the FGM table is based should resemble the characteristics of the multidimensional flame to yield an accurate representation of combustion chemistry. This chapter focusses on the basics of the FGM reduction method.

Throughout this dissertation, the Combustion Approximation is used in flamelets. All flamelets treated in this thesis are deflagrations for which the adoption of the low-Mach number approximation [Buckmaster1985], commonly referred to as the Combustion Approximation, is often used. By using low-Mach number asymptotes, it can be shown [Rook2001] that pressure fluctuations can be neglected compared to the ambient pressure. This implies that the pressure remains constant throughout the flamelet. Due to this approximation, all time- and spatial pressure derivatives drop out of equations (1.3) and (1.5) and the thermal equation of state, equation (1.11), reads:

$$\rho = p_\infty \overline{M} / (\mathcal{R} T)$$  \hspace{1cm} (2.1)

in which $p_\infty$ denotes the ambient pressure.

This chapter commences in section 2.1 the generic flamelet equations are deduced from the general set of conservation equations for reactive flow as described in section 1.2. In this thesis two types of flamelets will be used: (steady) premixed flamelets and (quasi-steady) counterflow diffusion flamelets. These two flamelet types represent the two combustion modes flames are commonly classified to: premixed and non-premixed combustion [Warnatz1996]. The reason for the use of these two flamelet types and their specific mathematical description is discussed in section 2.2. Section 2.3 explains how FGM tables are generated from individual flamelets.
2.1 Flamelet equations

A flame front can be treated macroscopically as an interface between unburnt and burnt mixture, but it can be treated microscopically as well, when the reaction layer is fully resolved. Section 2.1.1 treats the flame front macroscopically and serves as a starting point for section 2.1.2 which derives the equations that fully describe all species mass fraction profiles throughout a reaction-diffusion layer.

2.1.1 Flame front dynamics

In a spatial domain, a reactive layer can be identified by a scalar variable $S$ which obeys $\nabla S \neq 0$. This identification method requires a resolved reaction layer, i.e. no shock-like discrete interface. Reaction-diffusion layers are defined by an iso-surface of $S$, i.e. a surface at which $S(x,t)$ is constant. For premixed flames it is common to use a reaction progress variable $S = Y$ as principal controlling variable: this variable quantifies the transition from unburnt mixture ($S = Y_u$) to chemical equilibrium ($S = Y_b$). On the other hand, for non-premixed flames it is common to use the mixture fraction $S = Z$ as principal controlling variable to determine the local elemental composition between fuel ($S = Z_{fu}$) and oxidizer ($S = Z_{ox}$). In the subsequent derivation of the flamelet equations the distinction between premixed and non-premixed flames will not be made in order to keep the equations as general as possible. The kinematic equation describing the movement of scalar variable $S$ is given by

$$\frac{DS}{Dt} = \frac{\partial S}{\partial t} + u_f \cdot \nabla S = 0,$$

(2.2)

implying that the local value for $S$ can only change by the movement of the flame. The flame propagation velocity $u_f$ can be decomposed into the local fluid velocity $u$ and the local flame displacement velocity $s_D$:

$$u_f = u + s_D n,$$

(2.3)

which is represented graphically in figure 2.1. For passive scalars, which do not have a (chemical) source term and thereby no displacement velocity of their own, it is obvious that $s_D = 0$ must apply. $s_D$ is a field quantity which can vary due to flame stretch or equivalence ratio variations for example. The normal vector $n$ is defined as:

$$n = -\frac{\nabla S}{|\nabla S|},$$

(2.4)

which implies that the normal vector points towards the unburnt mixture for premixed flames provided that $Y_u < Y_b$. For non-premixed flames $n$ points towards the oxidizer side, provided that $Z_{ox} < Z_{fu}$. Substitution of equation (2.3) and (2.4) in equation (2.2) leads to:

$$\frac{\partial S}{\partial t} + u \cdot \nabla S = s_D |\nabla S|,$$

(2.5)

which is very similar to the kinematic $G$-equation which was introduced by Williams [Williams1985]. However, in the $G$-equation approach only a single iso-contour/interface $G = G^0$ is tracked, contrary to equation (2.5) in which $S$ is a continuous variable.
2.1 Flamelet equations

2.1.2 Generic flamelet equations

A generic system of one-dimensional flamelet equations can be derived from combustion equations (1.26-1.28) following the ideas of De Goey and Ten Thije Boonkkamp [Goey1997, Goey1999]. De Goey and Ten Thije Boonkkamp [Goey1997] defined a mass-based flame stretch rate $K$ of a fluid element moving with $u_f$ as:

$$K = \frac{1}{M} \frac{DM}{Dt} , \quad \text{with} \quad M (t) = \int_{V(t)} \rho \, dV ,$$

in which $M (t)$ represents the mass contained by a volume $V (t)$ in the flame. In figure 2.1 it is shown that the generic three-dimensional flame configuration can be reduced to a quasi one-dimensional structure having a curvilinear coordinate system with coordinate $s$ perpendicular to iso-surfaces of $S$, i.e. $e_s = -n$. Application of Reynolds Transport Theorem to $M (t)$ in equation (2.6) yields the following equation for the quantity $K$ in differential notation [Goey1997]:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho u_f) = \rho K .$$

Substitution of equation (2.3) and subtraction of the continuity equation (1.26), results in the mass conservation equation:

$$\nabla \cdot (\rho s_D n) = \rho K .$$

For a control volume on the curvilinear flame-adapted coordinate system with coordinate $s$, and by introducing the mass burning rate $m = \rho s_D$, equation (2.8) reduces to:

$$\frac{\partial m \sigma}{\partial s} = -\rho K \sigma ,$$

in which the minus-sign comes from definition (2.4) which reduces to $n = -1$ for a one-dimensional coordinate system. $\sigma$ is a measure for the flame surface on an iso-contour of $S$. For clarity, the $\nabla$-operator is consistently used when a multi-dimensional coordinate system is concerned, while the differential operator $\frac{\partial}{\partial s}$ is used for the one-dimensional coordinate system. Equation (2.9) can be interpreted as the transport equation for the amount of mass which is transported perpendicular to iso-contours of $S$ through $\sigma$.

One-dimensional equations are parameterized with the spatial coordinate $s$ in the curvilinear coordinate system, which is the arc-length perpendicular to the iso-contours of $S$. In this one-dimensional coordinate system, both $K$ and $\sigma$ are a function of coordinate $s$ and time only. The curvature $\kappa$ of iso-surfaces of $S$ is related to the derivative of $\sigma$ via
\[ \kappa = \nabla \cdot \mathbf{n} = -\frac{1}{\sigma} \frac{\partial \sigma}{\partial s}, \]  

(2.10)

and can be used to rewrite equation (2.9) to

\[ \frac{\partial m}{\partial s} = -\rho K + \kappa m. \]  

(2.11)

The generic transport equation for scalar \( S \), analogous to transport equation (1.28), reads:

\[ \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u} S) = \nabla \cdot \left[ \frac{\lambda}{L_p c_p} \nabla S \right] + \dot{\omega}_S. \]  

(2.12)

Substitution of equations (2.2), (2.3) and (2.7) yields the following expression:

\[ \nabla \cdot \left[ m S \mathbf{n} + \frac{\lambda}{L_p c_p} \nabla S \right] + \dot{\omega}_S = \rho K S, \]  

(2.13)

in which \( \mathbf{n} \) denotes the normal vector defined by equation (2.4). For a quasi one-dimensional configuration, equation (2.13) reduces to:

\[ \frac{\partial m \sigma S}{\partial s} - \frac{\partial}{\partial s} \left[ \sigma \frac{\lambda}{L_p c_p} \frac{\partial S}{\partial s} \right] = \dot{\omega}_S \sigma - \rho \sigma K S. \]  

(2.14)

By extraction of all flame curvature \( \kappa \) contributions, equation (2.14), can be restated as:

\[ \frac{\partial}{\partial s} (F_S) = \dot{\omega}_S - \rho K S + \kappa F_S, \]  

(2.15)

where \( F_S \) is the short-hand notations for the combination of the convective and diffusive flux of \( S \):

\[ F_S = mS - \frac{\lambda}{L_p c_p} \frac{\partial S}{\partial s}. \]  

(2.16)

Similar derivations as equation (2.15) can be made for one-dimensional conservation equations for species mass fractions and enthalpy which results in a system of equations, from now on referred to as flamelet equations:

\[ \frac{\partial m}{\partial s} = \kappa m - \rho K, \]  

(2.17)

\[ \frac{\partial F_S}{\partial s} - \dot{\omega}_S = \kappa F_S - \rho K S, \]  

(2.18)

\[ \frac{\partial F_i}{\partial s} - \dot{\omega}_i = \kappa F_i - \rho K Y_i + Q_i, \quad i \in [1, N_s - 1] \]  

(2.19)

\[ \frac{\partial F_h}{\partial s} = \kappa F_h - \rho K h + Q_h, \]  

(2.20)

in which all deviations from 1D flat flame behavior are gathered in the right-hand sides of the equations. Similar notations as in equation (2.16) are introduced for the convective and diffusive fluxes of \( Y_i \) and \( h \):

\[ F_i = mY_i - \frac{\lambda}{L_i c_p} \frac{\partial Y_i}{\partial s}, \quad i \in [1, N_s - 1] \]  

(2.21)

\[ F_h = m h - \frac{\lambda}{c_p} \frac{\partial h}{\partial s} - \frac{\lambda}{c_p} \sum_{i=1}^{N_s} \left( \frac{1}{L_i} - 1 \right) h_i \frac{\partial Y_i}{\partial s}. \]  

(2.22)
Transport equations (2.17-2.20) completely describe the internal structure of a flame. Compared to equation (2.15), additional terms occur in equation (2.19) and (2.20) due to the relative movement of of iso-surfaces of \( Y_i \) and \( h \) with respect to iso-surfaces of \( S \) [Oijen2002-2]. Misalignment of \( Y_i \) and \( h \) iso-surfaces with respect to \( S \) iso-surfaces can be caused by a difference in propagation velocity of \( Y_i \) and \( h \) with respect to \( S \), or by diffusive transport along the flame surfaces \( \sigma \) because iso-surfaces of \( Y_i \) and \( h \) generally do not coincide with iso-surfaces of \( S \). This introduces gradients in \( Y_i \) and \( h \) perpendicular to the normal \( n \); these gradients are denoted by the \( \nabla || \)-operator. The misalignment contributions are gathered in terms \( Q_i \) and \( Q_h \) which read:

\[
Q_i = \rho (u_{Y_i} - u_f) \cdot \nabla Y_i + \nabla \cdot \left( \frac{\lambda}{Le_i c_p} \nabla || Y_i \right), \quad i \in [1, N_s], \quad (2.23)
\]

\[
Q_h = \rho (u_h - u_f) \cdot \nabla h + \nabla \cdot \left[ \frac{\lambda}{c_p} \left( \nabla || h + \sum_{i=1}^{N_s} \left( \frac{1}{Le_i} - 1 \right) h_i \nabla || Y_i \right) \right], \quad (2.24)
\]

in which \( u_{Y_i} \) and \( u_h \) denote the propagation velocities of \( Y_i \) and \( h \) iso-surfaces respectively.

Up to now, no approximations are made in equations (2.17-2.20) describing the internal structure of a flamelet. However, if the flamelet is in (quasi-) steady state, the first term in \( Q_i \) and \( Q_h \) can be neglected. When all perturbations due to gradient in \( Y_i \) and \( h \) perpendicular to the flamelet curvilinear coordinate system are neglected, \( Q_i \) and \( Q_h \) vanish completely. Curvature effects are negligible when the curvature radius the flame surfaces \( \kappa^{-1} \) is much larger than the flame thickness \( \delta_L \); i.e. \( |\kappa^{-1}| \gg \delta_L \). In this thesis only flat, i.e. non-curved flamelets are considered, thereby dropping the curvature terms out of equations (2.17-2.20). The following system of one-dimensional equations remains:

\[
\frac{\partial m}{\partial s} = -\rho K \quad (2.25)
\]

\[
\frac{\partial mS}{\partial s} = \frac{\partial}{\partial s} \left( \frac{\lambda}{Le_S c_p} \frac{\partial S}{\partial s} \right) + \dot{\omega}_S - \rho KS \quad (2.26)
\]

\[
\frac{\partial mY_i}{\partial s} = \frac{\partial}{\partial s} \left( \frac{\lambda}{Le_i c_p} \frac{\partial Y_i}{\partial s} \right) + \dot{\omega}_i - \rho K Y_i, \quad i \in [1, N_s - 1] \quad (2.27)
\]

\[
\frac{\partial mh}{\partial s} = \frac{\partial}{\partial s} \left[ \frac{\lambda}{c_p} \frac{\partial h}{\partial s} + \frac{\lambda}{c_p} \sum_{i=1}^{N_s} \left( \frac{1}{Le_i} - 1 \right) h_i \frac{\partial Y_i}{\partial s} \right] - \rho K h. \quad (2.28)
\]

The internal flamelet structure is now defined in terms of species mass fractions \( Y_i(s) \), \( h(s) \) and mass burning rate \( m(s) \) for a stretch field \( K(s) \). The steady form of the equations shows that, within the moving flow field, quasi-steady flamelet structures can be identified which are transported by the flow field, but whose inner structure is not affected by flow motion. In general, stretch field \( K(s) \) cannot be found from a transport equation: it should be computed using its definition (2.6).

For both the mixture averaged viscosity \( \mu \) and the mixture averaged thermal conduction coefficient \( \lambda \) often empirical laws are used [Wilke1950, Mason1958, Mathur1967] to avoid the time-consuming computations required to evaluate these properties. In this thesis simplified formulations for \( \mu \) and \( \lambda \) of combustion gas mixtures will be used [Smook1991] to reduce computational cost:

\[
\frac{\mu}{c_p} = 1.67 \times 10^{-8} \left( \frac{T}{298} \right)^{0.51} \quad (2.29)
\]

\[
\frac{\lambda}{c_p} = 2.58 \times 10^{-5} \left( \frac{T}{298} \right)^{0.69} \quad (2.30)
\]
Bongers [Bongers2005-1] compared coefficients computed by equations (2.29) and (2.30) with predictions from more complex models [Wilke1950, Mathur1967] for CH$_4$-air and C$_{12}$H$_{26}$-air combustion, both at atmospheric and gas turbine conditions ($p = 25$ bar, $T_0 = 800$ K). It was found that for C$_{12}$H$_{26}$-air combustion the coefficients are more sensitive to pressure and temperature but values of coefficients remained within 5% of the values for atmospheric conditions.

2.2 Archetypical flamelet equations

In this thesis two specific cases of these flamelet types are used to generate FGM tables: steady premixed flamelets and quasi-steady counterflow diffusion flamelets. In a premixed flamelet, a stream of perfectly mixed fuel and oxidizer/air is converted to combustion products when it passes through the reactive layer. The system is treated as an adiabatic, freely-propagating, premixed flat flame. Combustion variables $\rho$, $T$, $T_i$ and the gas velocity only depend on the $x$-coordinate perpendicular to the flame surface. The flamelet is stretchless indicating that no mass is lost perpendicular to the flame coordinate system, i.e. $K = 0$. A schematic representation of a premixed flamelet is depicted in the left part of figure 2.2. In a counterflow flamelet a fuel jet impinges on an oxidizer/air jet forming a stagnation plane with a flat diffusion flame at, or close to, the stoichiometric plane. Since this flame type is considered to be flat, combustion variables $\rho$, $T$ and $T_i$ only depend on the $x$-coordinate perpendicular to the flame surface and time. The $x$-component of the gas velocity $u$ is a function of $x$-coordinate and time but the $y$-component of the gas velocity $v$ and pressure $p$ are functions of both coordinates $x$ and $y$ and time. A schematic representation of a counterflow diffusion flamelet is depicted in the right part of figure 2.2. Only planar, i.e. two-dimensional, counterflow geometries are considered in this thesis.

This section derives the equations that describe these two specific flamelet types, and applicable boundary conditions are given. The generic flamelet equations in section 2.1 were derived for a quasi-1D curvilinear coordinate system, in the following subsections Cartesian coordinates are used for convenience.
2.2 Archetypical flamelet equations

2.2.1 Steady stretchless premixed flamelet equations

For steady, stretchless premixed flamelets the combustion equations (1.26-1.29) reduce to:

\[
\frac{\partial \rho u}{\partial x} = 0 , \tag{2.31}
\]

\[
\frac{\partial \rho u Y_i}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda}{L_{ei} c_p} \frac{\partial Y_i}{\partial x} \right) + \dot{\omega}_i , \quad i \in [1, N_s - 1] \tag{2.32}
\]

\[
\frac{\partial \rho u h}{\partial x} = \frac{\partial}{\partial x} \left[ \frac{\lambda}{c_p} \frac{\partial h}{\partial x} + \frac{\lambda}{c_p} \sum_{i=1}^{N_s} \left( \frac{1}{L_{ei}} - 1 \right) h \frac{\partial Y_i}{\partial x} \right] , \tag{2.33}
\]

Keeping in mind that \( \partial x = \partial s \) and for a steady flame \( m = \rho u \), it is easily seen that this system of equations is equal to the flamelet equations (2.25-2.28) without stretch. Dirichlet, i.e. fixed-value, boundary conditions for \( Y_i \) and \( h \) are imposed at the unburnt side:

\[
Y_i (x \to -\infty) = Y_{i,-\infty} \quad \text{and} \quad h (x \to -\infty) = h_{-\infty} , \tag{2.34}
\]

which, for a given pressure, composition and temperature of fuel and oxidizer, can be described as a function of the flame stoichiometry \( \phi \) only. At the chemical equilibrium side, Neumann, i.e. zero-gradient, boundary conditions for \( Y_i \) and \( h \) are imposed:

\[
\frac{\partial Y_i}{\partial x} (x \to \infty) = 0 , \quad \frac{\partial h}{\partial x} (x \to \infty) = 0 . \tag{2.35}
\]

Boundary conditions for \( h \) can be replaced by \( T \) by using equation (1.9). The translational degree of freedom of the flame front is removed by appointing a fixed temperature at \( x = 0 \). The mass burning rate \( m = \rho u s_D = \rho u \) is an eigenvalue of equations (2.31-2.33); for adiabatic premixed flamelets, the flame displacement speed \( s_D \) is commonly referred to as the burning velocity \( s_L \). The flamelet equations can be solved by specialized one-dimensional flame codes like CHEM1D [Somers1994]. The unburnt mixture and the chemical equilibrium mixture represent the two extreme points in composition space which are linked by the solution of equations (2.31-2.33). This solution represents a one-dimensional curve in composition space, which can be regarded as a one-dimensional manifold.

Figure 2.3 depicts the typical spatial structure of a premixed flamelet. The upper left graph shows reactants, typical reaction products are shown in the upper right graph while intermediate species, the radicals formaldehyde and hydroxyl, are depicted in the lower left graph. In the lower right graph it can clearly be seen that a reactant, oxygen for example, is consumed (negative source terms) while a combustion product, carbon dioxide for example, is produced (positive source terms).

2.2.2 Unsteady counterflow diffusion flamelet equations

In the counterflow setup, the \( y \)-component of the flow is taken into account by the introduction of the local flame stretch rate \( G \) which is defined as [DixonLewis1990, Stahl1991]:

\[
G (x, t) \equiv \frac{\partial v}{\partial y} , \tag{2.36}
\]

and is a function of \( x \)-coordinate and time only. For steady counterflow flames it can be shown that \( G \) is equal to the mass-based flame stretch rate \( K \), see equation (2.6), but for
unsteady flames $G$ is more convenient to use. The profile for the flame stretch rate is still unknown but, contrary to $K$, the definition for $G$ allows the deduction of a transport equation for the local flame stretch rate. By substitution of equation (2.36), for a planar counterflow geometry, continuity equation (1.26) reduces to:

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho u}{\partial x} = -\rho G . \quad (2.37)$$

For the unknown flame stretch field $G$ a transport equation can be derived from the momentum equation (1.27) in $y$-direction [DixonLewis1990, Stahl1991]. This differential equation reads:

$$\rho \frac{\partial G}{\partial t} + \rho u \frac{\partial G}{\partial x} = \frac{\partial}{\partial x} \left( \mu \frac{\partial G}{\partial x} \right) + P(t) - \rho G^2 , \quad (2.38)$$

in which $P(t)$ denotes the pressure gradient in $y$-direction. The full derivation can be found in appendix A. In the far-field the flow is assumed to behave as a potential flow; in the oxidizer stream a velocity gradient can now be prescribed which only depends on time:

$$\frac{\partial u}{\partial x}(x \to \infty) = -\frac{\partial v}{\partial y} = -G = -a(t) , \quad (2.39)$$

in which $a(t)$ denotes the applied strain rate which can be a function of time. By means of substitution of equation (2.39) in equation (2.38), for $P(t)$ is obtained:

$$P(t) = \rho_{ox} \left( \frac{\partial a(t)}{\partial t} + a(t)^2 \right) . \quad (2.40)$$
For unsteady counterflow diffusion flamelet combustion equations (1.26-1.29) can now be rewritten as:

\[
\frac{\partial \rho}{\partial t} + \frac{\partial \rho u}{\partial x} = -\rho G, \tag{2.41}
\]

\[
\frac{\partial \rho h}{\partial t} + \frac{\partial \rho u h}{\partial x} = \frac{\partial}{\partial x} \left( \mu c_p \frac{\partial h}{\partial x} \right) + \rho_{\text{ox}} \left( \frac{\partial a(t)}{\partial t} + a(t)^2 \right) - \rho G^2, \tag{2.43}
\]

Reminding that \( \partial x = \partial s \) and \( m = \rho u \), flamelet equations (1.26-1.29) are recovered with additional unsteady terms. Dirichlet boundary conditions for \( Y_i \) and \( h \) are imposed in both the oxidizer \( (x \to -\infty) \) and fuel \( (x \to \infty) \) stream:

\[
Y_i(x \to \infty) = Y_{i,\text{ox}}, \quad h(x \to \infty) = h_{\text{ox}}, \tag{2.45}
\]

\[
Y_i(x \to -\infty) = Y_{i,\text{fu}}, \quad h(x \to -\infty) = h_{\text{fu}}, \tag{2.46}
\]

while for \( G \) a combination of Dirichlet and Neumann boundary conditions is imposed:

\[
G(x \to \infty) = a(t), \quad \frac{\partial G}{\partial x}(x \to -\infty) = 0. \tag{2.47}
\]

The translational degree of freedom is removed by choosing the stagnation plane \( u = 0 \) at \( x = 0 \). The parameter of this system is the applied strain rate \( a \) which is defined at the oxidizer side \( (x \to \infty) \). Steady counterflow diffusion flamelet solutions can be obtained for \( a \leq a_{\text{ext}} \) where \( a_{\text{ext}} \) denotes the extinction strain rate. For \( a > a_{\text{ext}} \) heat and (radical) species are removed from the reaction zone too fast; as a consequence chemical kinetics become too slow to burn the reactants leading to quenching of the flamelet. The (theoretical) lower limit for \( a \) equals zero which corresponds to chemical equilibrium everywhere in the flamelet. Contrary to premixed flamelets, counterflow diffusion flamelets do not possess a fixed mass burning rate \( m \) or flame thickness \( \delta_L \). Besides the fuel and oxidizer composition and temperature, \( m \) and \( \delta_L \) strongly depend on \( a \). For counterflow diffusion flamelets \( \delta_L \) can be estimated by: \( \delta_L \propto \sqrt{D/a} \) in which \( D \) denotes the diffusion coefficient. This correlation implies that \( \delta_L \) increases rapidly for small values of \( a \) making a numerical solution procedure very sensitive to variations in \( a \). Therefore, a small value \( (a = 0.25 \text{ [1/s])} \) is chosen as minimal value for \( a \), which corresponds to a composition close to chemical equilibrium.

Figure 2.4 depicts the generic spatial structure of a steady counterflow diffusion flamelet for \( a = 100 \text{ s}^{-1} \). The upper left graph shows reactants, typical reaction products are shown in the upper right graph while intermediate species, the radicals formaldehyde and hydroxyl, are depicted in the lower left graph. In the lower right graph it can clearly be seen that a reactant, oxygen for example, is consumed (negative source terms) while a combustion product, carbondioxide for example, is produced (positive source terms). The most obvious distinction from premixed flamelet structures, as shown in figure 2.3, is that fuel and oxidizer gradients are opposed for counterflow diffusion flamelets while they are aligned for premixed flamelets.
2.3 FGM construction for partially-premixed applications

In this section it is described how a manifold is generated from the one-dimensional flame equations. This approach is slightly different for manifolds generated from premixed flamelets, described in section 2.2.1, and counterflow diffusion flamelets, described in section 2.2.2. In section 2.3.1 the independent variables used to parameterize manifolds are defined. Subsequently, the procedures to generate manifolds from flamelets are clarified in section 2.3.2. Finally, in section 2.3.3 it is explained what operating procedure is applied when an FGM is used to represent combustion chemistry in a flame simulation.

2.3.1 FGM parametrization

The full composition space of an isobaric reactive system comprises $N_s + 1$ dimensions: $Y_i$ and $T$. The chemical manifold is a low-dimensional subspace of this composition space; when the dimensionality of the manifold is increased a larger part of the full composition space can be accessed by the manifold. Increasing the dimensionality can, theoretically, be continued up to $N_s + 1$ dimensions: in this case the full composition space is recovered. An analysis of DNS data by Maas and Thevenin [Maas1998-1] indicated that the minimum dimension of an ILDM manifold, defined by the maximum error in species mass fraction predictions, remained significantly smaller than the full composition space dimensionality. Similar results have been obtained in other studies concerning ILDM tables [Schmidt1999] and FGM tables [Oijen2007] as well. At the same time, extending the dimensionality of a database also implies that more differential equations have to be solved and a more complex look-up procedure is to be used, resulting in a reduced gain in computational time.

In reality, it is not straightforward to define multiple control variables to parameterize a combustion chemistry table. Ideal control variables are independent of one another and
give single-valued functions for all dependent variables. When control variables can not be considered to be fully independent, the requirement should be met that subsequent control variables exhibit a monotonous behavior when conditioned on the previous ones. For the FGM reduction method, as well as for the purely mathematical reduction methods ILDM [Maas1992] and CSP [Goussis1994], both the number of control variables is user-defined and definition of the control variables onto which the manifold is projected. This is an intuitive approach since the physical interpretation of control variables is straightforward and remains constant over the entire manifold. However, the use of fixed control variables can introduce manifold subspaces where the manifold is ill represented by the control variables, i.e. the control variables are quasi-constant while dependent variables do change significantly or the control variables do not yield a unique representation of the manifold. To overcome these numerical issues, the Generalized Coordinates [Bauer2006, Maas1998-2] method has been introduced which locally adapts the parametrization such that it follows the manifold. This method uses the eigenvectors associated with the smallest negative eigenvalues of the chemical system as control variables, which change over the manifold. This results in control variables that give a unique representation of the manifold and are ideally adapted to track reaction progress. However, since the parametrization is defined during generation of the manifold, combination with the FGM reduction method is not straightforward. On top of that, the use of generalized coordinates erases the physical meaning of control variables. For these reasons the conventional approach, in which user-defined control variables are used to parameterize the manifold, is employed in this thesis. For partially-premixed combustion at least two control variables are needed: one to describe mixing of fuel and oxidizer and one to describe reaction progress from unburnt to chemical equilibrium.

Mixture fraction

Because fuel and oxidizer are not perfectly mixed in partially-premixed flames, element mass fractions $Z_k$ change due to mixing. As pointed out in section 1.2, chemical reactions can not change $Z_k$: they merely recombine these elements into other molecules. In partially-premixed flames, the local equivalence ratio, which is determined by the local element composition, is commonly described by the mixture fraction $Z$. By defining $Z$ as a function of element mass fractions, it becomes a conserved scalar, unaffected by chemical kinetics. Bilger [Bilger1990] defined an element-based, expression for the mixing variable $Z$:

$$Z = \frac{2 M_H^{-1} [Z_H - Z_{H,2}] + 0.5 M_C^{-1} [Z_C - Z_{C,2}] - M_O^{-1} [Z_O - Z_{O,2}]}{2 M_H^{-1} [Z_{H,1} - Z_{H,2}] + 0.5 M_C^{-1} [Z_{C,1} - Z_{C,2}] - M_O^{-1} [Z_{O,1} - Z_{O,2}]},$$

(2.48)

in which $Z_k$ denotes the molar mass of an element $k$ and subscripts $H$, $C$, $O$, 1 and 2 refer to hydrogen, carbon, oxygen and the fuel- and oxidizer stream, respectively. $Z$ is a normalized quantity which distinguishes quantitatively between oxidizer ($Z = 0$), fuel ($Z = 1$) and intermediate states. For flames in which preferential diffusion is absent, $\phi$ is directly related to $Z$ by

$$\phi (Z) = \frac{sZ}{(1-Z) Y_{O_2,ox}},$$

(2.49)

in which $s$ denotes the stoichiometric mass ratio of oxidizer and fuel ($s = 3.989$ for methane-air flames; $Y_{O_2,ox} = 0.233$ for air).
Reaction progress variable

For the conversion of unburnt to burnt mixture it is convenient to define a reaction progress variable $Y$ [Bray2005] which discriminates quantitatively in the transition from unburnt mixture to the chemical equilibrium composition. $Y$ must be monotonous from the initial (unburnt) state to chemical equilibrium in both lean and rich regions in order to facilitate an unambiguous mapping of dependent variables. In non-premixed flames a classic choice for the indicator of reaction progress is the scalar dissipation rate $\chi$ [Peters1984, Veynante2002], which is proportional to the squared absolute mixture fraction gradient. The positive-definite value $\chi$ loosely translates to the strain rate $a$ in counterflow diffusion flamelets: increasing values for $\chi$ correspond to a composition further away from chemical equilibrium in a similar way as increasing $a$-values do in counterflow diffusion flamelets. In this thesis FGM tables based on both premixed as counterflow diffusion flamelets are used and therefore a reaction progress indicator is required which is defined in both types of flamelets, preferably with a direct link to the formation of reaction products. A linear combination of species mass fractions has the required properties for the reaction progress variable $Y$:

$$Y = \sum_{i=1}^{N_s} \alpha_i Y_i,$$

in which $\alpha_i = 0$ for all species except for CO\textsubscript{2}, H\textsubscript{2}O and H\textsubscript{2} is applied. Weight factors for these species are $\alpha_{\text{CO}_2} = M_{\text{CO}_2}^{-1}$, $\alpha_{\text{H}_2\text{O}} = M_{\text{H}_2\text{O}}^{-1}$ and $\alpha_{\text{H}_2} = M_{\text{H}_2}^{-1}$ which ensures a monotonous behavior from the unburnt mixture to chemical equilibrium for methane-air flames between flammability limits. For the definition of $Y$ given by equation (2.50), $Y_0 = 0$ for any given value of $Z$.

Flamelet solutions can now be mapped on $Z$ and $Y$, viz. $\varphi = \varphi (Z, Y)$ in which $\varphi$ can denote any thermochemical variable. The feasibility of the $Z$-$Y$ approach has been demonstrated by several others [Oijen2004, Pierce2004, Pitsch2000, Vreman2008]. A two-dimensional manifold already requires a considerable amount of computer memory but, when sufficient computer memory is available, the resolution and dimensionality of FGM tables can straightforwardly be increased for more accuracy. As an example, non-adiabatic effects due to radiative heat transfer or flame stabilization are not captured by this parametrization. An additional control variable, $h$ for example [Oijen2000], would be required for an adequate description of these heat loss mechanisms.

2.3.2 FGM generation

Over the years, several tabulation methodologies have been developed. Tabulation of combustion chemistry can be done during run-time by means of the In Situ Adaptive Tabulation (ISAT) [Pope1997] or Piecewise Reusable Implementation of Solution Mapping (PRISM) [Tonse1999] do. The advantage of these methods is that they only compute and store thermochemical variables in the accessed composition space effectively keeping computer memory requirements low. In a priori tabulation approaches, a choice has to be made on which reduction method the combustion chemistry table is based. Mathematical reduction methods like ILDM [Maas1992], CSP [Goussis1994] and REDIM [Bykov2006] are available, or flamelet-based methods like FGM [Oijen2000] and FPI [Gicquel2000] can be opted for. Tabulation, both during run-time and a priori and independent of the chemistry representation method (detailed/mathematically reduced/flamelet), can be replaced by...
the use of Artificial Neural Networks [Blasco1999, Christo1996]. They allow the reduction of required computer memory by several orders of magnitude [Ihme2009], but their performance fully depends on their (assumed) structure and the amount and accuracy of the data which is used to train them [Ihme2008-1]. In this thesis, equidistantly spaced structured look-up tables are used and thermochemical variables are retrieved using (bi-)linear interpolation. This method is preferred because of its simplicity, robustness and speed: the equidistant spacing allows direct pinpointing of the table entries which are required for interpolation. Structured tables require a larger amount of memory, since also the composition space which is not accessed is tabulated. Methods like the high-order orthogonal polynomial parameterizations [Turanyi1994] have been proposed to reduce the computer memory requirements of structured tables.

FGM tables from premixed flamelets

To generate a two-dimensional FGM from premixed flamelets, the flamelet equations described in section 2.2.1 are solved for multiple, different $Y_i$ and $h$ boundary conditions at $x \to -\infty$. These variables are directly related to $Z$ according to

$$Y_i (x \to -\infty) = Z Y_{i, fu} + (1 - Z) Y_{i, ox}, \quad h (x \to -\infty) = Z h_{fu} + (1 - Z) h_{ox},$$

or can be related to $\phi$ through equation (2.49). Flamelet solutions are computed and subsequently stored as a function of the control variables $Z$ and the normalized progress variable $C$. Since the equilibrium composition, and thereby the progress variable equilibrium value $Y_{eq}$ depends on $Z$, $C$ is introduced to enable efficient look-up procedures:

$$C = \frac{Y - Y_0 (Z)}{Y_{eq} (Z) - Y_0 (Z)} \in [0, 1].$$

For the definition of $Y$ given by equation (2.50), $Y_0 = 0$ for any given value of $Z$. Each previous flamelet serves as an initial guess of the new solution, and since only small variations in the boundary conditions are made, convergence is reached within a few iterations. For isobaric combustion, the composition of the chemical equilibrium mixture is
completely determined by the values of the conserved variables \( Z_k \) and \( h \) and is computed and added to each flamelet. The left part of figure 2.5 depicts such a series of stretchless, steady premixed flamelet solutions for different \( Z \)-values. From the flamelet solutions, \( \dot{\omega}_i \), \( \dot{\omega}_y \), \( \dot{\omega}_i \) and \( T \) are interpolated on an equidistant \( Z-C \) grid. \( \rho \) and \( c_p \) are subsequently recalculated to keep them consistent with \( Y_i \) and \( T \). When the table has to be extended beyond the flammability limits, for which no steady solution of equations (2.31-2.33) exists, linear interpolation is applied between the leanest flamelet and the oxidizer composition, viz.

\[
\varphi (Z,C) = \left[ \frac{Z}{Z_{\text{fl}}} \right] \varphi (Z_{\text{fl}},C) + \left[ \frac{Z_{\text{fl}} - Z}{Z_{\text{fl}}} \right] \varphi_{\text{ox}}, \quad Z \in [0,Z_{\text{fl}}],
\]

(2.53)

and between the richest flamelet and the fuel composition, viz.

\[
\varphi (Z,C) = \left[ \frac{Z - Z_{\text{ufl}}}{1 - Z_{\text{ufl}}} \right] \varphi_{\text{fu}} + \left[ \frac{1 - Z}{1 - Z_{\text{ufl}}} \right] \varphi (Z_{\text{ufl}},C), \quad Z \in [Z_{\text{ufl}},1],
\]

(2.54)

in which \( Z_{\text{fl}} \) and \( Z_{\text{ufl}} \) denote the lower and upper flammability limit in terms of \( Z \) and \( \varphi \) can denote any thermochemical variable. Figure 2.6 depicts \( T \) and \( \dot{\omega}_y \) from an example of a final FGM table. In previous simulations of confined laminar triple flames [Oijen2004], FGM tables based on premixed flamelets gave a very accurate representation of combustion chemistry with only a fraction of the computational effort required for detailed chemistry.

FGM tables from counterflow diffusion flamelets

Since counterflow diffusion flamelets extinguish at a limiting value for the applied strain rate, the composition subspace between the most strained steady counterflow diffusion flamelet solution and the mixing solution can not be covered if only steady counterflow diffusion flamelet solutions are used. To tackle the issue of a natural continuation beyond the extinction limit, the unsteady formulation of the flamelet equations is used. A proposed model is the Lagrangian Flamelet Model (LFM) [Pitsch2000] in which case the Lagrangian flamelet time is a control variable. Since the specification of this parameter is far from trivial in general cases, this method is not used here. In the method proposed in this thesis, the applied strain rate is a function of time, e.g. \( a = a(t) \) and a small value \((a = 0.25 \text{ [1/s]})\) is chosen as initial value. The strain rate is subsequently increased in time.
2.4 Summarizing overview

In this chapter the concept of the Flamelet Generated Manifold (FGM) reduction method has been introduced. The FGM method assumes that the flame structure in two or

using an exponential function with a timescale that is significantly larger than that of the slowest evolving species: it is verified that quasi-steady counterflow diffusion flamelet solutions having a strain rate below the extinction strain rate are (nearly) identical to their steady flamelet solutions. The time-dependent solution of equations (2.41-2.44) is tracked until the flame is extinguished and the solution equals the mixing state. The right part of figure 2.5 depicts such a series of unsteady counterflow diffusion flamelet solutions for different \( a \)-values. Subsequently, \( Y_i \) and \( T \) from the flamelet solutions are interpolated on an equidistant \( Z-C \) grid. Like for the premixed flamelet-based FGM table, \( \rho \) and \( c_p \) are recomputed for consistency with \( Y_i \) and \( T \). The chemical equilibrium mixture, which is a function of conserved variables \( Z_k (Z) \) and \( h (Z) \) and thereby \( Z \) only for isobaric combustion, can be added to the flamelet solutions provided that \( Y \) remains monotonous for every \( Z \)-value. Figure 2.7 depicts \( \rho \) from two different final FGM table; the most visible distinction at \( Z = 1 \) originates from the difference in boundary conditions for premixed and counterflow diffusion flamelets.

2.3.3 FGM usage

After the FGM table is generated, it can be linked to a CFD code. This implies that the CFD code has to solve transport equations for controlling variables \( Z \) and \( Y \) together with continuity and momentum equations (1.26-1.27). No additional energy equation has to be solved since \( h \) and \( T \) are functions of \( Z \) and \( Y \) only. The differential equation for \( Z \) is derived by taking the proper linear combination of \( Y_i \):

\[
\frac{\partial \rho Z}{\partial t} + \frac{\partial \rho u_j Z}{\partial x_j} - \frac{\partial}{\partial x_j} \left( \frac{\lambda}{c_p} \frac{\partial Z}{\partial x_j} \right) = \frac{\partial}{\partial x_j} \left( \frac{\lambda}{c_p} \sum_{i=1}^{N_a} a_i \left( \frac{1}{Le_i} - 1 \right) \frac{\partial Y_i}{\partial x_j} \right),
\]

(2.55)

in which \( a_i \) follows from the mixture fraction definition (2.48) and equation (1.25). From equation (2.55) it can clearly be seen that \( Z \) is not influenced by chemical reactions but merely by advection and diffusion of species. Application of equation (2.50) over equation (1.28) yields the transport equation for \( Y \):

\[
\frac{\partial \rho Y}{\partial t} + \frac{\partial \rho u_j Y}{\partial x_j} - \frac{\partial}{\partial x_j} \left( \frac{\lambda}{c_p} \frac{\partial Y}{\partial x_j} \right) - \sum_{i=1}^{N_a} \alpha_i \lambda = \frac{\partial}{\partial x_j} \sum_{i=1}^{N_a} \alpha_i \left( \frac{1}{Le_i} - 1 \right) \frac{\partial Y_i}{\partial x_j},
\]

(2.56)

where \( \alpha_i \) denotes the weight factors defined in section 2.3.1. In the case of unit Lewis numbers the entire right-hand side of equations (2.55) and (2.56) vanish. During a flame simulation, \( Y \)-values from the CFD solver are scaled to \( \mathcal{C} \) whenever \( \rho, \omega_Y, T \) and \( c_p \) are retrieved from the FGM database. The replacement of an individual transport equation for each species by equations (2.55) and (2.56) results in a large reduction in both the number of equations and the stiffness of the system of equations. Equations (2.55) and (2.56) do include multi-dimensional perturbations due to flame stretch and curvature and, if detailed chemistry would be used, these perturbations would affect thermochemical variables. However, with the current parametrization the FGM does not account for their influence on the correlation of thermochemical variables. Additional control variables can be used to account for these effects, see [Swart2010, Vreman2009-2] for example, but this is beyond the scope of this research.

2.4 Summarizing overview

In this chapter the concept of the Flamelet Generated Manifold (FGM) reduction method has been introduced. The FGM method assumes that the flame structure in two or
three-dimensional flames resembles the structure of one-dimensional flames, i.e. the gradient in thermochemical variables in the principal direction is a few orders of magnitude than the gradient in directions perpendicular to the principal direction. This implies that the multi-dimensional flame structure can accurately by one-dimensional transport equations containing convection, diffusion and chemical reaction contributions. Like other manifold methods, the FGM method can be used to generate a multi-dimensional chemical manifold, whereas a single flamelet can only be considered as a one-dimensional manifold. Ergo, the FGM method spans a subspace of the entire composition space while methods based on a single flamelet only contain a single trajectory in composition space. The FGM reduction method can therefore be considered as a combination of classic flamelet- and manifold approaches. Compared to conventional mathematical reduction methods like ILDM [Maas1992] and CSP [Goussis1994], the FGM method has proven to be as accurate close to chemical equilibrium but significantly more accurate in (cold) regions where chemistry is dominated by transport of species [Oijen2000], i.e. where chemical reactions are balanced by convection and diffusion.

The implementation of the FGM method in CFD codes is straightforward: thermochemical variables from flamelets are stored in an FGM table prior to the CFD simulation. During the simulation, differential equations are solved for the controlling variables and required thermochemical data like chemical source terms and density is retrieved from the FGM table by means of interpolation. The use of transport equations for the controlling variables implies that controlling variables are defined in the entire computational domain. Local flame front deformation in terms of stretch and curvature are thereby resolved and their influence on e.g. reaction rates can be encompassed by the FGM table, in contrary to level-set methods like the $G$-equation approach [Williams1985]. The associated drawback is that the reactive layer has to be resolved, which requires a high spatial resolution of the numerical grid. For flame simulations in which full resolution of the reactive layer is impossible due to numerical restrictions, simulation strategies to overcome this restriction will be discussed in chapter 5.

Compared to ILDM and CSP, the FGM method has no general procedure for the generation of FGM tables. Although it can be automated to a large extend, the user still has
to determine the type, *i.e.* premixed or counterflow diffusion, and boundary conditions of
flamelets which are used to construct the FGM table. In chapter 3 stratified flames are
investigated for which previous research has pointed out that premixed flamelets are most
applicable to generate the FGM table. In chapters 4 and 5, for the well-known
partially-premixed Sandia Flames it is examined which type of flamelet is most
appropriate and subsequently an FGM table based on this flamelet type is used in Large
Eddy Simulations of the moderately turbulent Flame D and the heavily turbulent Flame F.
DNS of stratified Bunsen flames

In this chapter Direct Numerical Simulations of stratified flames are performed, in which FGM tables are used to keep computational costs within acceptable limits. The small to moderate stratifications, keeping the entire flame within flammability limits, make it an obvious choice to use premixed flamelets to generate the FGM table. The stratified flames are analyzed and it is assessed whether the Flame Surface Density (FSD) model could be used to replace the current approach: the advantage of the FSD model is that the reaction layer no longer needs to be resolved. In the FGM approach, the reaction layer is required to be resolved which leads to high numerical requirements.

3.1 Introduction

Flames observed in many engineering appliances are stratified, i.e. quasi-premixed with a range in equivalence ratio $\phi$ present at the flame front. Examples of these appliances are gas turbine combustors, spark ignition reciprocating engines and industrial furnaces. Operational behavior of these combustors is extensively analyzed and optimized by numerical simulations in order to reduce cost and design time. The unsteady nature of these turbulent flames requires time-dependent simulations when detailed information on flame characteristics are needed. Over the last decade, the Large Eddy Simulation (LES) approach has proven to become feasible for simulation of industry-relevant problems. In LES of these flames, the flame thickness is generally smaller than the numerical filter size; therefore the reactive layer can not be resolved and subfilter influences have to be taken into account by a model. On the other hand, Direct Numerical Simulations (DNS) are often prohibitive even for moderate Reynolds numbers due to their high computational cost. DNS is a valuable tool for development of subfilter models for LES applications but the method is too expensive for application in a design environment.

An approach to work around the issues associated with the non-resolvedness of the reactive layer in premixed combustion is the Artificially Thickened Flame (ATF) approach [Colin2000]. In this approach the diffusion coefficient(s) are multiplied with a factor $\alpha > 1$ and the chemical source term(s) are divided by the same factor $\alpha$ which results in the same mass burning rate [Poinsot2001]. The ratio between the flame thickness and the turbulent length scale is affected resulting in a different turbulence-chemistry interaction; for this reason the ATF approach is abandoned in this study. The G-equation model [Peters2000] assumes an infinitely thin flame sheet which is only influenced by
advection and flame propagation. It has the advantage that its model is relatively simple but the disadvantage of this method is that the variable G is only defined at the flame sheet contrary to progress variable approaches. Progress variable values, and derived quantities like flame surface densities, are defined in the entire computational domain and can be extracted from DNS or experimental measurements.

In this chapter it is examined whether models based on the Flame Surface Density (FSD) approach [Marble1977, Pope1988, Candel1990], which has been developed for turbulent premixed combustion, can also be applied to stratified flames. The concept of the FSD model is that the volumetric production of reaction progress variable is replaced by an infinitely thin flame sheet, like the G-equation model, in which the unburnt mixture is converted to burnt instantaneously. The filtered molecular transport and chemical source terms are jointly modeled as a source term which is proportional to a mass burning rate \( m \) and the subfilter flame surface density \( \Sigma \) which represents the amount of flame surface per unit of volume. For stratified flames it is investigated whether the FSD model can be applied and how the variation in mass burning rate should be taken into account.

This study will continue from the turbulent premixed planar \( \text{CH}_4 \)-air Bunsen flames simulated by Vreman et al. [Vreman2009-1], which are similar to flames studied by Filatyev et al. [Filatyev2005], Bell et al. [Bell2007] and Sankaran et al. [Sankaran2007]. The novelty of this work is the introduction of a stratification and the objective of this study is to perform an a priori analysis of the performance of the FSD combustion model for stratified flames based on DNS data. Since the main envisioned application is a gas turbine combustor, ranges of \( \phi \) as observed in these combustors are applied. To reduce the computational requirements for a DNS to an acceptable level, the Flamelet Generated Manifold (FGM) method [Oijen2000], also known as the Flamelet Prolongated ILDM (FPI) method [Gicquel2000], is used to reduced combustion chemistry. The FGM reduction method generates a manifold based on one-dimensional flame structures and can therefore be considered to be a combination of classic flamelet- and manifold methods. A database is created for specific well-defined conditions without the removal of elementary reactions or species from the used chemical reaction mechanism. In this database thermochemical variables are stored as a function of a small number of rate-controlling variables for which a transport equation is solved. This results in a large reduction in the number of required transport equations and a reduction of the stiffness of the system of equations. FGM tables composed of steady premixed flamelets have proven to be accurate for premixed Bunsen-type flames including heat loss effects [Oijen2000], highly stretched premixed counterflow flames [Oijen2002-1] and confined triple flames [Oijen2004]. In DNS of an expanding turbulent flame using the FGM method to represent combustion chemistry [Oijen2005], the use of a single control variable gave accurate predictions of the local mass burning rate. Additional control variables can be used to increase the accuracy of the FGM method.

Stratified methane-air flames are treated here: the assumption of unit Lewis numbers is appropriate to these fuel-air mixtures which allows the simplification of the transport equations. Law and Sung [Law2000] state that the Lewis number of the deficient species, i.e. the fuel in lean mixtures, is the mixture’s effective Lewis number. The Lewis number of \( \text{CH}_4 \) is close to unity for conditions observed in the current study, e.g. 0.992 for a premixed flamelet with \( \phi = 0.7 \) and \( T_0 = 800 \text{ K} \) at atmospheric pressure. For the globally lean \( \text{CH}_4 \)-air flames treated in this chapter, the unit Lewis number assumption will most
3.1 Introduction

probably not cause major deviations from results that would be obtained experimentally.

It is subsequently assumed that thermochemical parameters can be mapped on only two independent control variables: the mixture fraction $Z$ describing mixing and the reaction progress variable $Y$ describing reaction progress, viz. $\varphi = \varphi(Z, Y)$ in which $\varphi$ can denote any thermochemical variable. The feasibility of this approach has been demonstrated by others [Pierce2004, Pitsch2000, Vreman2008]. Four different turbulent flames are simulated: one premixed flame which serves as a reference case and three stratified flames. In these stratified flames the effect of stratification is increased both by an increase in equivalence ratio gradient and an increase in equivalence ratio range. This allows the separation of effects caused by the stratification itself and the effects caused by indirect effects like an extended range of the mass burning rate, which depends heavily on $Z$. This study will focus on modeling of the mass burning rate for stratified flames, not on modeling of the flame surface density $\Sigma$. Several types of models for $\Sigma$ have been proposed by others: algebraic models [Boger1998], similarity models [Knikker2002] and models involving the solution of a transport equation for $\Sigma$ [Boger1998, Weller1998, Hawkes2000]. A simple algebraic model for $\Sigma$, in essence the Bray-Moss-Libby (BML) model, is used to complete the FSD-model; for more complex models for $\Sigma$ the reader is referred to literature. For the stratified flames in this study it is examined whether a subfilter Dirac- or $\beta$-PDF for the filtered mass burning rate yields accurate predictions. This analysis is done for multiple realistic LES filter widths.

Chapter structure

Section 3.2 describes the DNS code which is used to generate resolved flame data from which the flame surface is extracted. This includes the equations in the DNS code, their numerical discretization methods and how the used FGM database is generated. Four different turbulent flames are simulated: the entire numerical set-up and boundary conditions for these DNS are described in section 3.3. In this section results from the DNS are also presented to obtain a good impression of the character of the simulated flames: flame structure, flame curvature and flame stratification will be discussed in detail. Section 3.5 subsequently describes the filtering method and results from a priori analysis of the FSD model for filter widths equal to two, four and eight times the reference flame thickness. Simultaneously, it will be examined whether subgrid variations in chemical source terms of rate-controlling variables can be accurately modeled using a combined PPDF for the control variables of the FGM database. Finally, conclusions from this study are summarized and discussed in section 3.6.
3.2 Used DNS solver and FGM generation

DNS equations and numerical discretization

For both the control variables $Z$ and $Y$ unit Lewis numbers are assumed. The Navier-Stokes equations for turbulent combustion with tabulated chemistry then read:

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho u_j}{\partial x_j} = 0,$$

$$\frac{\partial \rho u_i}{\partial t} + \frac{\partial \rho u_i u_j}{\partial x_j} = \frac{\partial p}{\partial x_i} + \frac{\partial}{\partial x_j} \left[ \mu \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) - \frac{2}{3} \mu \frac{\partial u_k}{\partial x_k} \delta_{ij} \right],$$

$$\frac{\partial \rho Z}{\partial t} + \frac{\partial \rho u_i Z}{\partial x_j} = \frac{\partial}{\partial x_j} \left( \frac{\lambda}{c_p} \frac{\partial Z}{\partial x_j} \right),$$

$$\frac{\partial \rho Y}{\partial t} + \frac{\partial \rho u_i Y}{\partial x_j} = \frac{\partial}{\partial x_j} \left( \frac{\lambda}{c_p} \frac{\partial Y}{\partial x_j} \right) + \dot{\omega}_Y,$$

where the summation convention is used over the indices $j$ and $k$ and a Newtonian behavior of the fluid is assumed. $\rho, \dot{\omega}_Y, T$ and $c_p$ are retrieved from the FGM table using linear interpolation in both $Z$ and $Y$ direction. Flame stretch and curvature are included in the DNS since transport equations are solved for $Z$ and $Y$, but a two-way coupling between stretch and curvature and the chemical source term is absent. Simplified formulations for $\mu$ and $\lambda$ for CH$_4$-air flames are used [Smooke1991] to reduce computational cost:

$$\mu/c_p = 1.67 \times 10^{-8} (T/298)^{0.51},$$

$$\lambda/c_p = 2.58 \times 10^{-5} (T/298)^{0.69}.$$

A standard finite volume method is employed on a staggered Cartesian mesh. For the momentum equations second-order central differencing and an explicit hybrid timestepping method are applied: convective terms are integrated in time using a third-order Adams-Bashforth scheme while for viscous terms a forward Euler scheme is used. The hybrid timestepping method is formally only first-order accurate but provides significantly better stability than a pure Adams-Bashforth or a pure forward Euler method. The contribution, and thereby also the error contribution, of the viscous terms scales with the inverse of the Reynolds number when compared to the convective terms. Therefore, for turbulent flames as considered in this study in which $Re = O(10^2)$, the viscous terms can be discretized using a lower order method than the convective terms without sacrificing numerical accuracy. For the scalars $Z$ and $Y$ the Van Leer's third-order accurate MUSCL scheme is applied to the advective terms while second-order central differencing is applied to the viscous terms. The variable density approach, which involves a Poisson equation for the pressure, has been successfully applied by Pitsch and Steiner [Pitsch2000] and Pierce and Moin [Pierce2004]. The multigrid implementation in the code used for this study is thoroughly described in [Vreman2009-1]. The code has been parallelized using both the MPI and OpenMP protocol.

FGM generation

For stratified flames the question arises whether premixed or diffusion flamelets have to be used to generate the FGM. Both Bongers et al. [Bongers2005-2] and Fiorina et al. [Fiorina2005-1] independently showed that when $\phi$ ranges between zero and
3.2 Used DNS solver and FGM generation

approximately two, a steady, stretchless premixed flamelet-based FGM describes partially-premixed flames better than a counterflow diffusion flamelet-based one. Therefore, in this study steady, stretchless premixed flamelets will be used to construct the FGM database. An *a posteriori* analysis of DNS data will be discussed in section 3.3 to verify whether the choice to use steady premixed flamelets is a valid one. Unit Lewis numbers are assumed for all species for consistency with the DNS code. The equations describing steady premixed flamelets with unit Lewis numbers for all species have been derived in section 2.2.1:

\[
\frac{\partial \rho u}{\partial x} = 0 ,
\]

\[
\frac{\partial \rho u Y_i}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda}{c_p} \frac{\partial Y_i}{\partial x} \right) + \dot{\omega}_i , \quad i \in [1, N_s - 1] ,
\]

\[
\frac{\partial \rho h}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda}{c_p} \frac{\partial h}{\partial x} \right) ,
\]

\[
\rho = p_{\infty} M / (R T) ,
\]

in which the low Mach-number approximation is used in the thermal equation of state (3.10) with \( p_{\infty} \) denoting the ambient pressure. \( h \) and \( c_p \) are tabulated polynomial functions of \( T \) [Kee1986]. The same simplified formulation for \( \lambda \), equation (3.6), as used in DNS equations (3.1-3.4) is used to close the flamelet equations. Source terms for species are computed using the GRI 3.0 reaction scheme [GRImech30]; the mass fraction of the abundant species \( N_2 \) is determined using the identity \( \sum Y_i = 1 \). All flamelet computations have been performed with the in-house flame code Chem1D [Somers1994] and the chemical equilibrium is explicitly added to each flamelet to ensure the correct equilibrium composition for slow evolving species. Besides variables that are changed by chemical reactions, the mass burning rate \( m = \rho_0 s_L \) is stored for every flamelet.

Flamelet solutions are computed and stored as a function of the control variables \( Z \) and \( Y \). \( Z \) is extracted from the flamelets using the definition based on element composition by Bilger [Bilger1990]:

\[
Z = \frac{2 M_H^{-1} [Z_H - Z_{H,ox}] + 0.5 M_C^{-1} [Z_C - Z_{C,ox}] - M_O^{-1} [Z_O - Z_{O,ox}]}{2 M_H^{-1} [Z_{H, fu} - Z_{H, ox}] + 0.5 M_C^{-1} [Z_{C, fu} - Z_{C, ox}] - M_O^{-1} [Z_{O, fu} - Z_{O, ox}]},
\]

in which \( Z \) denotes element mass fractions of respectively hydrogen \( H \), carbon \( C \) and oxygen \( O \). The subscripts \( fu \) and \( ox \) denote the fuel (\( CH_4 \)) and oxidizer (air) respectively. For the reaction progress variable \( Y \) a linear combination of species mass fractions is used:

\[
Y = \sum_{i=1}^{N_s} \alpha_i Y_i ,
\]

in which \( \alpha_i = 0 \) for all species except for \( CO_2 \), \( H_2O \) and \( H_2 \). Weight factors for these species are \( \alpha_{CO_2} = M_{CO_2}^{-1} \), \( \alpha_{H_2O} = M_{H_2O}^{-1} \) and \( \alpha_{H_2} = M_{H_2}^{-1} \) which allows unambiguous mapping of the dependent variables for the \( CH_4 \)-air flames in the considered range of \( \phi \). This definition for \( Y \) has been applied with success in previous studies [Oijen2002-2, Vreman2008]. Since the equilibrium composition, and thereby the progress variable equilibrium value \( Y_{eq} \), depends on \( Z \), a normalized progress variable \( C \), defined by

\[
C = \frac{Y - Y_0 (Z)}{Y_{eq} (Z) - Y_0 (Z)} \in [0, 1] ,
\]
is introduced. For the definition of \( Y \) given by equation (3.12), \( Y_0 = 0 \) for any given value of \( Z \). \( Y_i \) and \( T \) from the flamelets are subsequently interpolated on an equidistant \( Z-C \) grid and \( \rho \) and \( c_p \) are recalculated using equation (3.10) and the previously mentioned polynomials in \( T \) to keep them consistent. During a DNS \( Y \) is scaled to \( C \) whenever \( \rho, \dot{\omega}_Y, T \) and \( c_p \) are retrieved from the FGM database and passed to the flow solver.

For the presentation of results, \( \phi \) is used instead of \( Z \) in this paper. For \( CH_4 \)-air flames \( \phi \) is related to \( Z \) by

\[
\phi (Z) = \frac{sZ}{(1-Z)Y_{O_2,2}}
\]  
(3.14)

in which \( s \) denotes the stoichiometric mass ratio of oxidizer and fuel (\( s = 3.989 \) for \( CH_4 \)-air flames); \( Y_{O_2,2} = 0.233 \) for air.

### Flame surface extraction

To validate the FSD approach, the flame surface area has to be extracted from DNS data. The flame surface is defined here as an iso-\( C \) surface. A surface integration method by Geurts [Geurts2001] is used for both surface determination and integration of thermochemical variables over the flame surface. This method is second-order accurate like the spatial discretization of the DNS code; it was verified by comparing the predicted surface area for perfectly spherical flame kernels with their analytical surface area. Flame kernels had different radii and different flame thicknesses in which the transition from \( C = 0 \) to \( C = 1 \) is governed by a hyperbolic tangent function. These results comply with a study by Bastiaans et al. [Bastiaans2009] in which this surface integration method was also used.

For the flames discussed in this paper the iso-\( C \) contour at which \( \dot{\omega}_Y \) achieves its maximum value is chosen. For the premixed flame with \( \phi = 0.7 \) this occurs at \( C = 0.711 \) as is shown in figure 3.1. Since the \( C \)-value at which the maximum \( \dot{\omega}_Y \) is reached is a function of \( \phi \), the averaged \( C \)-value at which the maximum \( \dot{\omega}_Y \) is obtained will be used for the stratified flames. For a stratification of \( 0.4 \leq \phi \leq 1.0 \) the average value for \( C \) at which the \( \dot{\omega}_Y \) achieves its maximum equals \( C = 0.727 \) while for \( 0.1 \leq \phi \leq 1.3 \) the average value for \( C \) equals \( C = 0.767 \). Results are not very sensitive to this choice.
3.3 DNS setup for stratified planar Bunsen flames

A central planar jet, having a width of $W = 2.4$ mm, consists of preheated unburnt mixture ($T_0 = 800$ K, $C = 0$) and has a streamwise mean velocity of $\bar{w}_0 = 20$ m/s. This mean velocity corresponds to a jet Reynolds number of $Re_{jet} = 607$. The co-flow surrounding the central planar jet, consisting of the same gas mixture in chemical equilibrium state ($C = 1$), has a streamwise mean velocity of 5 m/s. A stratification is applied along the slot (in the periodic direction) for both the central jet and co-flow by a prescribed mixture fraction gradient. The inflow plane is depicted schematically in figure 3.2 ($z = 0$).

Four different turbulent Bunsen flames are simulated: one premixed reference case denoted by TP and three stratified flames having the same mean equivalence ratio ($\bar{\phi} = 0.7$ or $Z = 3.93 \cdot 10^{-2}$) but different stratifications. The stratification is introduced by a mixture fraction variation with amplitude $\Delta_Z$ is given by:

$$ Z(x) = Z_M + \Delta_Z \cos \left( \frac{2\alpha \pi x}{L_x} \right) $$ (3.15)

in which $L_x$ denotes size of the computational domain in the periodic direction. In flame TSI one period of the $Z$-perturbation corresponds to $L_x$, i.e. $\alpha = 1$. Flame TSII is identical to flame TSI except for the doubled wave number, i.e. $\alpha = 2$ which is applied to increase the gradient in $\phi$. For both flame TSI and TSII the stratification corresponds to $0.4 \leq \phi \leq 1.0$. Flame TSIII is identical to flame TSI except that the stratification in $Z$ corresponds to $0.1 \leq \phi \leq 1.3$. These values are chosen as such to introduce a reduction in

---

**Figure 3.2:** Boundary conditions in the numerical setup: $W (= 2.4 \text{ mm})$ denotes the slot width. The cosine indicates the stratification in the inlet plane. The dark grey plane represents the central planar jet of unburnt mixture with a streamwise mean velocity of $\bar{w}_0 = 20$ m/s while the light grey plane represents the co-flow of burnt mixture with a streamwise mean velocity of $\bar{w}_0 = 5$ m/s. In the $yz$-plane an instantaneous contour of the flame front at $x = 1.25W$ is shown.
the mixing length scale similar to flame TSII but to introduce larger (non-linear) variation in the mass burning rate as well. Turbulent inflow conditions are generated using a random noise generator: for each velocity component, every timestep random numbers are generated on the mesh one grid cell level below the inflow plane. A box-filter of $\Delta_f = 0.25W$ in all three spatial directions and a corresponding temporal filter are subsequently applied; the temporal filter is discretized by

$$\hat{f}_{n+1} = \left( 1 - \frac{\nu_0 \Delta t}{\Delta_f} \right) \hat{f}_n + \left( \frac{\nu_0 \Delta t}{\Delta_f} \right) f_n$$

(3.16)

The perturbation is initialized with $\hat{f}_0 = 0$ and multiplied by an appropriate amplification factor to obtain the required inflow turbulence intensities.

A rectangular computational domain, shown in figure 5.4, is used in which the planar jet lies parallel to the $x$-axis; the centerline of the jet corresponds to $y = 0$. A uniform grid spacing of $\Delta x = 23.4 \mu m$ is used which was stretched in the far field in $y$-direction ($|y|/W \geq 0.8$). The transition between the central jet and co-flow in terms of both progress variable and streamwise velocity is described by a hyperbolic tangent function with a length scale equal to the flamelet flame thickness $\delta_0$. This transition zone enforces flame stabilization at the inflow plane. $\delta_0$ is based on the temperature increase and the maximum temperature gradient for a premixed, preheated CH$_4$-air flamelet with an equivalence ratio of $\phi = 0.7$; $\delta_0$ equals 0.329 mm which corresponds to $14 \Delta x$. For the stratified flames the transition length scale was kept equal to $\delta_0$ for all $x$-coordinates, i.e. the transition length is not adjusted to local $\phi$-values. It is assumed that the flame structure will evolve rapidly from the imposed hyperbolic tangent function to its structure governed by combustion chemistry when convected downstream. Periodic boundary conditions are applied in $x$-direction for velocity components, pressure and transported scalars. In both $y$-direction and the outflow-plane in $z$-direction Neumann (zero-gradient) boundary conditions are applied for velocity components and transported scalars. Besides the periodic pressure boundary conditions in $x$-direction, a Dirichlet ($p = p_0$) boundary condition is applied in $y$-direction and a Neumann boundary condition for the pressure is applied at the inflow and outflow plane. A constant time step of $\Delta t = 7.5 \times 10^{-2} \mu s$ is used for all simulations in order to keep the CFL-number sufficiently low. An overview of simulation parameters and turbulent flame characteristics can be found in table 3.1.

To generate the required FGM tables, 1201 preheated, premixed CH$_4$-air flamelets are computed between $\phi = 0.1$ and $\phi = 1.3$ using an equidistant step in $\phi$. All flamelets have an initial temperature $T_0 = 800$ K and a constant ambient pressure $p_0 = 101.325$ kPa. For flame TP only the premixed flamelet with $\phi = 0.7$ is stored to reduce required memory. For flame TSI and TSII an FGM with an equidistant $201 \times 201 Z-C$ grid is generated from the corresponding $0.4 \leq \phi \leq 1.0$ flamelets. For flame TSIII an FGM with an equidistant $401 \times 201 Z-C$ grid is compiled to assure that the interpolation error in $Z$-direction is comparable for all stratified flames. As an example, FGM entries for temperature and progress variable source term for flame TSIII are shown in figure 3.3; the FGM table for flames TSI and TSII is a subset of the shown table.

Section 3.4 will discuss results from the, fully resolved, DNS data. All flames will be characterized by relevant dimensionless numbers and flame thickness, flame curvature and stratification at the flame surface will be examined in depth. This is done to get a thorough impression of the flame character and to verify that chemistry representation by premixed flamelets has been an appropriate choice.
3.4 Results and analysis of resolved turbulent stratified flames

To verify the DNS code, a premixed laminar Bunsen flame is simulated first. Identical jet and co-flow velocities are used as in the turbulent flames but the turbulent perturbations at the inlet are omitted; this results in an unperturbed V-shaped Bunsen flame. An equivalence ratio of \( \phi = 0.7 \) is used for which the flame propagation velocity equals \( s_0^{\phi} = 1.434 \text{ m/s} \) and the laminar mass burning rate equals \( m_0 = \rho_0 s_0^{\phi} = 0.611 \text{ kg/(m}^2\text{s)} \). The mass consumption rate \( m \) in the laminar Bunsen flame is computed by

\[
m = \frac{m_0}{\phi}
\]
\[ m A_f = \int_{A_{in}} \rho w [1 - C] \, dA \]  

(3.17)

in which \( A_f \) denotes the surface of an iso-\( C \) contour and \( A_{in} \) denotes the inflow plane. Flame surfaces are defined as iso-\( C \) contour; values for \( C \) have been defined in section 3.2.

The mass consumption rate obtained from this premixed laminar Bunsen flame simulation is only 2.5% larger than the laminar mass burning rate \( m_0 \) of the corresponding premixed flamelet: \( m = 0.626 \text{ kg/(m}^2\text{s)} \). This difference is attributed to the choice for the value of \( C \) at which the flame surface is taken, and thereby the flame surface \( A_f \). The influence of flame tip curvature on the chemical source term is also not accounted by the currently used FGM parametrization. For the arbitrary choice of \( C = 0.85 \) the error of the predicted mass burning rate reduces to 0.06% but no physical argument can be given for this value of \( C \). For the remainder of this study, the \( C \)-value discussed in section 3.2 will be used to define the flame surface location.

### 3.4.1 Stratified flame statistics

The Kolmogorov length scale \( \eta \), streamwise Taylor length scale \( \lambda_T \) and integral length scale \( L_{INT} \) are defined by

\[ \eta = (\nu^3/\epsilon)^{1/4} \]  

(3.18)

\[ \lambda_T = \sqrt{\frac{w'w'/(\partial w' / \partial z)^2}{}} \]  

(3.19)

\[ L_{INT} = k^{3/2}/\epsilon \]  

(3.20)

in which \( \nu \) denotes the kinematic viscosity, \( k \) denotes the turbulent kinetic energy, \( \epsilon \) denotes the viscous dissipation and \( w' \) denotes the streamwise velocity fluctuation. Time or volume averaged quantities are denoted by an overbar, e.g. \( \overline{\varphi} \), while surface averaged quantities will be denoted by single guillemets, e.g. \( \langle \varphi \rangle \). For filtering purposes, 15 realizations are stored for each case. These realizations are separated in time by \( \Delta t = 0.75 \) ms: since the average throughflow time of the turbulent flames equals 0.88 ms, all realizations are assumed to be statistically independent. Time-averaging of velocity and scalar samples was started at \( t = 1.8 \) ms which corresponds to approximately two throughflow times. Statistics of velocity and scalar quantities were computed at \( t = 7.5 \) ms and these results were compared to results obtained after 15 realizations (\( t = 11.25 \) ms) to verify that the statistics were converged. Instantaneous iso-surfaces of the flame surface are shown for flame TSI in figure 3.4 and flame TSII in figure 3.5.

Table 3.1 shows that \( \eta > \Delta x = 23.4 \mu m \) and that length scales \( \eta, \lambda_T \) and \( L_{INT} \) are similar for all flames. The length scales and dimensionless numbers are extracted at the jet centerline plane \( (y = 0) \), one slot width downstream of the inflow plane \( (z = W) \). It is concluded that the structure of turbulence is similar for all turbulent flames. The Damkohler number, defined as \( Da = (\lambda_T/w') / (b_0/s_0^0) \), is slightly below unity for all turbulent flames. Rutland and Trouvé [Rutland1993] state that when \( Da > 1 \) the local flame structure remains laminar: the nearly-unity Damkohler numbers indicate that the flames will behave laminar-like. The Karlovitz number, \( Ka = (\delta/\eta)^2 \), is well below 100 for all turbulent flames; according to Peters [Peters2000] and based on the observed Damkohler and Karlovitz numbers, the turbulent flames are in the Thin Reaction Zones (TRZ) regime. In the next subsection it is verified that the flames are in the TRZ regime by looking at the flame structure in more detail.
3.4 Results and analysis of resolved turbulent stratified flames

3.4.2 Flame thickness

The local flame structure can be compared with flame structures observed in one-dimensional laminar flames by looking at the magnitude of the progress variable gradient. Protruding turbulent eddies and flame front curvature may cause local flame thickening which is indicated by a lower progress variable gradient than observed in laminar one-dimensional flames. Progress variable gradients are computed in the entire computational domain using the same central finite-difference second-order scheme as used in the DNS code. Subsequently, results are conditioned on $C$ and stored in 30 bins. For the stratified flames results are conditioned on $\phi$ as well: bins in $\phi$-space were defined as $\Delta\phi = 0.01$ around the mean bin value. Figure 3.6 displays that for $\phi = 0.7$ conditional

*Figure 3.4: Subsequent flame surface snapshots of flame TSI separated by 0.75 ms which corresponds to 85% of one throughflow time. Two flames are superposed in periodic direction; the surface color indicates the $\phi$-value at the iso-surface ranging from $\phi = 0.4$ to $\phi = 1.0$. Flame surfaces are defined by the average progress variable $C$ at which the maximum $\dot{\omega}_Y$ is obtained; for this case $\bar{C} = 0.727$. 


mean gradients observed for both flame TP and TSI resemble conditional mean gradients observed in premixed flamelets very well. The mean flame structure is not influenced by turbulence or the applied stratification. The error bars indicate that turbulent effects, both the intrusion of small eddies and local dilatation, deform the preheat zone but do not influence the reaction layer too much. These results comply well with the conclusions drawn from the dimensionless numbers in table 3.1; for flames TSII and TSIII very similar results were obtained (not shown). When regions towards the lean and stoichiometric limit in the stratified flames are considered, the mean flame structure in the DNS computations still resembles premixed flamelet structures very well, as is shown in figure 3.7 for flame TSI at $\phi = 0.5$ and $\phi = 0.9$. Intrusion of turbulent eddies and dilatation can still only affect the preheat zone, not the reaction layer. The flamelet thickness equals 0.420 mm for $\phi = 0.5$ and 0.304 mm for $\phi = 0.9$ which explains the lower maximum progress variable gradient.
for $\phi = 0.5$. For these values of $\phi$, flames TSII and TSIII again showed very similar results for the flame structure (not shown). The only deviations from the flamelet structure were observed in very lean regions ($\phi = 0.2$) close to the inlet plane where the reaction layer profile has to develop from the prescribed hyperbolic tangent profile with length scale $\delta_0$, as depicted in the left graph of figure 3.8. Because of very small values of $\dot{\omega}_Y$ in these lean regions, the reaction layer profile can not swiftly adapt to the flamelet profile. For $\phi = 1.2$ the turbulent flame can adapt to the flamelet profile much faster due the large values of $\dot{\omega}_Y$. Therefore no deviation from the flamelet profile is observed on the rich side.

Besides the confirmation that the flames are really in the TRZ regime, which is of interest for many engineering appliances, it also shows that the flames are still in the flamelet regime for which the FGM reduction method for reaction kinetics is applicable.

### 3.4.3 Flame front curvature

The influence of flame curvature and stretch on $\dot{\omega}_Y$ is not accounted for by the used FGM parametrization. Curvature radii in the turbulent flames should thus be significantly larger than the flame thickness, i.e. $\kappa^{-1} \gg \delta_0$, to be able to justify the exclusion of curvature effects in the FGM database. To allow the verification whether this exclusion is justified, flame front curvature radii are extracted from DNS data and compared to $\delta_0$. The mean flame front curvature $\kappa$ is obtained by computing the divergence of the normal vector of the flame front $n_F$, defined by $n_F = \nabla Y / |\nabla Y|$. By definition $n_F$ points towards the burnt mixture and is only defined when $\nabla Y \neq 0$. For more detail on the flame front curvature, the two local principal flame front curvatures $\kappa_1$ and $\kappa_2$ can be obtained by determining the two non-zero eigenvalues of the curvature tensor $n_{ij}$ [Dopazo2006]

$$n_{ij} = |\nabla Y|^{-1} \sum_{k=1}^{3} \left( \delta_{ik} - \frac{\partial Y}{\partial x_i} \frac{\partial Y}{\partial x_k} \right) \frac{\partial^2 Y}{\partial x_j \partial x_k}.$$  

(3.21)

The advantage of the latter method is that saddle points can be distinguished from non-curved flame fronts for which both $\kappa = \kappa_1 + \kappa_2 \approx 0$. In saddle points both eigenvalues are non-zero and have a comparable absolute value but one is positive and the other negative, i.e. $\kappa_1 \approx -\kappa_2$ while $|\kappa_1| \gg 0$. For non-curved flame fronts, both eigenvalues are negligible. A convenient characterization of the flame front curvature was introduced by Pope et al. [Pope1989] by the definition of the curvature shape factor $\mathcal{H}$:

$$\mathcal{H} = \begin{cases} \frac{\kappa_1}{\kappa_2} & \text{for } |\kappa_2| \geq |\kappa_1|, \\ \frac{\kappa_2}{\kappa_1} & \text{for } |\kappa_1| > |\kappa_2|. \end{cases}$$

(3.22)

Using definition (3.22), saddle points correspond to $\mathcal{H} = -1$ and a spherically curved flame front corresponds to $\mathcal{H} = 1$. For $\mathcal{H} = 0$ one principal curvature can be neglected compared to the other one indicating a cylindrically curved flame surface. Points having $\kappa_i < \kappa_j < 1.0 \times 10^{-6}$ are removed for this analysis: the ratio of $\kappa_i$ and $\kappa_j$ might, misleadingly, indicate cylindrical flame structures while in reality the flame front is practically flat.

The PDF of flame curvature is shown in the left graph of figure 3.9 for flames TP and TSIII. For flame TP, the obtained mean flame front curvature probability profile and the curvature shape factor probability profile are very similar to results from another premixed turbulent Bunsen flame reported by Bell et al. [Bell2007]. Surface averaged normalized flame front curvatures $\langle \kappa_0 \rangle$ range between 0.150 for flame TSII to 0.162 for
flame TSIII which corresponds to a length of approximately 0.9W. Since the flame remains attached at the burner slot edges these values for $\langle \kappa \rangle$ are to be expected for this Bunsen flame slot width. More than 70% of the flame surface has a mean curvature radius which is larger than the flame thickness, this corresponds to the region $P (-1 \leq \kappa \delta_0 \leq 1)$ indicated by the vertical dashed lines in the left part of figure 3.9. Smaller curvature radii, i.e. higher $|\kappa|$-values, come from turbulent eddies which are able to intrude the inner layer. For flame TSIII, only 1.3% of the flame surface has a mean curvature radius smaller than $\delta_0/3$, for the other flames this value is even smaller. Flame TSIII exhibits a slightly larger probability of
3.4 Results and analysis of resolved turbulent stratified flames

The right graph of figure 3.9 shows that the probability of occurrence of (spherical) saddle points ($\mathcal{H} = -1$) is the same for all flames but the probability of occurrence of cylindrically curved flame surfaces ($\mathcal{H} = 0$) tends to increase when $\Delta \phi$ is increased. Apparently, the increase in stratification tends to increase the probability of occurrence of flat or cylindrically curved flame surfaces. No significant difference in either mean flame front curvature or curvature shape factor distribution was observed between flame TSI and TSII indicating that the higher stratification, enforced by the double wave number for the periodic $Z$ function at the inlet, does not significantly affect the flame curvature. Flame TSIII exhibits a higher probability of occurrence of cylindrically curved flame surfaces. This is probably caused by the small Damkohler number in regions with small $\phi$-values: here the high streamwise gas velocity is able to stretch the flame surface in streamwise direction. The very lean (blue) regions in figures 3.4 and 3.5 do exhibit such a streamwise stretching.

It can be concluded that the numerical grid is fine enough to capture flame curvatures present in these Bunsen flames. The observation that for over 70% of the flame surface $\kappa^{-1} > \delta_0$ confirms that nearly no eddies small enough to perturb the reaction layer exist. Saddle points, identified by $\kappa \approx 0$ and $\mathcal{H} \approx -1$, are an important curvature morphology but cylindrically curved deformations, identified by $\kappa \neq 0$ and $\mathcal{H} \approx 0$, are preponderant.

3.4.4 Stratification at the flame surface

Previous sections led to the conclusions that the simulated Bunsen flames are all in the TRZ regime and that the assumption to neglect flame curvature effects on reaction kinetics is a suitable one. The last issue to be addressed in this section is the mixing length scale $L_{\text{mix}} = |\nabla \phi|^{-1}$ observed in the stratified flames. The FGM database is generated from premixed flamelets which allow diffusion only in $Y$-direction: no interaction between individual flamelets, or diffusion in $\phi$-direction, is taken into account. This assumption is valid only when the mixing length is significantly larger than the flame thickness, i.e. $L_{\text{mix}} \gg \delta_0$. In this section it is examined whether the choice to use premixed flamelets to
generate the FGM database is appropriate from a mixing perspective. Keeping the adoption of the FSD model in mind, a premixed combustion mode should be observed to justify the application of the FSD model which is intended for premixed combustion.

The probability density function of $\phi$ at the flame surface is extracted from flames TSI, TSII and TSIII and is shown in the left graph of figure 3.10. The surface averaged equivalence ratio $\langle \phi \rangle$ is smaller than 0.7 for all three flames: $\langle \phi \rangle = 0.674$ for TSI, $\langle \phi \rangle = 0.690$ for TSII and $\langle \phi \rangle = 0.608$ for TSIII. The, strictly taken, non-linear relation between $Z$ and $\phi$ given by equation (3.14), has only a very minor influence on the deviation of the $\langle \phi \rangle$ from the value 0.7. For $P(Z) = 1$, $\int \phi(Z) P(Z) dZ = 0.697$ for flames TSI and TSII and 0.687 for flame TSIII. Hence this non-linearity can not cause the large deviation of $\langle \phi \rangle$ from the value 0.7 for flames TSI and TSIII. The large probability of flame surface with a low $\phi$-value in flames TSI and TSIII is caused by the smaller Damkohler numbers at these $\phi$-values, allowing the flame surface to be stretched more in the streamwise direction. In flame TSIII the left peak of the PDF is shifted slightly more towards $\phi = 0.7$ than in flame TSI because the low Damkohler numbers allow more mixing. Application of the double wave number of the stratification is an efficient way to enhance mixing as can be observed when $P(\phi)$ of flame TSIII is compared to that of flame TSI. The PDF of $\phi$ exhibits a less bimodal behavior for flame TSI than for flame TSI and, as a consequence, the variance of the $\phi$-PDF of flame TSIII is smaller than for flame TSI.

The mixing length scale $L_{\text{MIX}}$ is extracted from DNS data using the fact that $\phi$ is a function of $Z$ only, given by equation (3.14):

$$\nabla \phi = \nabla Z \frac{\partial \phi}{\partial Z}(Z),$$  \hspace{1cm} (3.23)

in which $\nabla Z$ is extracted using a central second-order scheme and $\partial \phi/\partial Z(Z)$ can be deduced analytically from equation (3.14). The right graph of figure 3.10 displays the PDF of the mixing length scale for all three stratified flames. The surface averaged mixing length scale $\langle L_{\text{MIX}} \rangle$ decreases from 16.0$\delta_0$ to 12.8$\delta_0$ when the wave number of the stratification is doubled, e.g. when flames TSII is compared to flame TSI. The mean $\langle L_{\text{MIX}} \rangle$ even decreases to 10.1$\delta_0$ when $\Delta \phi$ is doubled, e.g. when flames TSIII is compared to flame TSI. The mixing length scale having the maximum probability of occurrence equals 9.7$\delta_0$, 6.1$\delta_0$ and 4.8$\delta_0$ for flame TSI, TSII and TSIII, respectively. For all flames the mean value for $\langle L_{\text{MIX}} \rangle$ is one order of magnitude larger than $\delta_0$ and even the mixing length scale having the maximum probability of occurrence is at least five times $\delta_0$ implying that the choice to use premixed flamelets to generate the FGM table is an appropriate one.

The PDF of the dot product of $n_\phi = \nabla \phi / |\nabla \phi|$ and $n_C = \nabla C / |\nabla C|$ at the flame surface, depicted in figure 3.11, shows a deviation from a random distribution: preference for alignment of the $n_\phi$ and $n_C$ vectors appears for all flames. In figure 3.11 only results for flame TSIII are shown but the other two stratified flames show the same behavior, albeit less apparent. This preference tends to increase with increasing $\Delta \phi$ and seems insensitive to the wave number of the stratification. The most straightforward explanation for this observation is that the surface averaged equivalence ratio $\langle \phi \rangle$ is smaller than the $\int \phi(Z) dZ$ value, or in other words, a large part of the flame surface is leaner-than-average. The probability to encounter a higher $\phi$-value in the burnt side of the flamelet is higher than the probability to encounter an even leaner mixture. This preference of alignment was also observed by Richardson et al. [Richardson2010] and they have shown that this alignment can give rise to “back supported” flamelets having a higher mass burning rate than flamelets in which $\phi$ remains constant. Since combustion chemistry for the DNS
3.5 Subfilter analysis

In LES applications, equation (3.4) is replaced by its spatially filtered version. Here, a top-hat filter with length scale $\Delta_t$ is used, which is denoted with an overbar. For the velocity components and progress variable the common Favre-Average (FA) is used, i.e. $\bar{\varphi} = \bar{p} \varphi / \bar{p}$. A subfilter viscosity model is implemented and the FSD model can be used to replace the entire right-hand side of the filtered equation (3.4) [Boger1998]:

$$\frac{\partial \bar{\varphi}}{\partial t} + \frac{\partial \bar{u}_j \bar{\varphi}}{\partial x_j} = \frac{\partial}{\partial x_j} \left[ \left( \frac{\lambda}{c_p} + \frac{\mu_t}{Sc_t} \right) \frac{\partial \bar{\varphi}}{\partial x_j} \right] + \bar{\omega} \bar{\varphi} \approx \langle m \rangle \left[ \bar{\varphi}_0 (Z) - \bar{\varphi}_e (Z) \right] \Sigma,$$

(3.24)

in which $\mu_t$ denotes the turbulent viscosity and $\Sigma$ denotes the flame surface density. $\langle m \rangle$ denotes the flame surface averaged mass burning rate. Since the progress variable $\bar{\varphi}$ is converted from $\bar{\varphi}_0$ to $\bar{\varphi}_e$ during combustion, contrary to the mass fraction of burnt mixture which ranges between 0 and 1, the source term in equation (3.24) contains a
multiplication with \([\mathcal{Y}_{eq} - \mathcal{Y}_0]\). Depending on the definition of \(\mathcal{Y}\), both \(\mathcal{Y}_{eq}\) and \(\mathcal{Y}_0\) can be a function of \(Z\). With the current definition for \(\mathcal{Y}\), given by equation (3.12), \(\mathcal{Y}_0\) is equal to zero for any \(Z\)-value and will therefor not be mentioned further. The mass burning rate \(m\) of stretchless premixed flamelets is shown in figure 3.13: it shows that the entire range from a negligible mass burning rate to its maximum value is present in flame TSIII and, to a lesser extend, in flames TSI and TSI. The maximum value for \(m\) occurs at \(\phi = 0.911\) and equals \(1.12 m_0 = 0.684 \text{ kg/(m}^2\text{s})\). For stoichiometric and rich mixtures the mass burning rate using unit Lewis numbers deviates from the mass burning rate using complex molecular transport but, as mentioned in section 3.1, in this study unit Lewis numbers are assumed to keep computational cost within limits.

Questions to be addressed are whether the FSD combustion model is suitable to stratified flames and, if applicable, how the variation in local mass burning rate should be accounted for. In this section it is assessed how well the FSD model can reproduce filtered chemical source terms for both the premixed and the stratified turbulent Bunsen flames. For LES of premixed flames, the filter size \(\Delta_f\) is typically up to one order of magnitude larger than the flame thickness. Therefore, in this analysis values for \(\Delta_f\) equal to \(\delta_0, 2\delta_0, 4\delta_0\) and \(8\delta_0\) are used. Figure 3.14 shows that all turbulent flames exhibit a very similar parabolic profile for \(\Sigma\) as a function of the FA normalized progress variable \(\bar{C}\), indicating that modeling of \(\Sigma\) might be very similar for premixed and stratified flames treated in this study.

In section 3.5.1 and 3.5.2, \(\Sigma\) is extracted from DNS data using Geurts’ method, which was discussed in section 3.2. In section 3.5.3, \(\Sigma\) is replaced by a simple algebraic model for \(\Sigma\).
More sophisticated models for $\Sigma$ are assumed to improve FSD results even more.

### 3.5.1 Flame surface analysis over the complete domain ($\Delta_F \rightarrow \infty$)

The FSD model assumes an infinitely thin flame sheet, which will result in deviations from finite-rate chemistry models when $\Delta_F$ become small. For small values of $\Delta_F$, the filter volume might contain a non-negligible chemical source term while no flame surface is identified resulting in an underestimation of the chemical source term by the FSD model. On the other hand, the filter volume might contain the flame sheet surface but not the entire flame thickness resulting in an overestimation of the chemical source. To exclude these effects, first the FSD model is validated over the entire computational domain. The flame surface area is extracted at iso-$C$ contours mentioned in section 3.2. For each element of this surface the local mass burning rate $m = m(\phi(Z); x)$ is retrieved from the flamelet database. From the DNS data a volume averaged source term and a flame surface density source term can be extracted which are defined by:

\[
\bar{\omega}_Y = \frac{1}{V_{\text{tot}}} \int_{V_{\text{tot}}} \dot{\omega}_Y(Z, Y; x) \, dV, \tag{3.25}
\]

\[
\langle m \rangle \mathcal{Y}_{\text{eq}}(Z) \Sigma = \frac{1}{A_f} \int_{A_f} m(\phi(Z); x) \mathcal{Y}_{\text{eq}}(Z) (Z; x) \, dA. \tag{3.26}
\]

Identical realizations as mentioned in section 3.4.1 are used for filtering. Figure 3.15 proves that the FSD assumption holds well for both the premixed flame and all stratified flames when the local mass burning rate $m(\phi(Z); x)$ is used in the surface integral in equation (3.26).

### 3.5.2 Modeling the surface averaged mass burning rate $\langle m \rangle$

The FSD model for stratified flames requires the surface averaged mass burning rate $\langle m \rangle$ in which subfilter variations, due to equivalence ratio variations, are accounted for. To obtain filtered data that represent LES results, DNS data is averaged using a box-filter with a width of $\Delta_F$. FA means and variances are computed for $Z$ and $C$ over each filter volume. It is examined whether these fluctuations can adequately be modeled using a presumed PDF to model subfilter variations in $m$. A $\beta$- and (Dirac) $\delta$-PDF are proposed as presumed models.
FIGURE 3.14: Scatter plots of the filtered flame surface density $\Sigma$ as function of the Favre-averaged normalized progress variable $\tilde{C}$ for the turbulent flames. For all plots the filter width $\Delta_f$ equals $4\delta_0$. The solid line represents a fit of a parabolic function.

FIGURE 3.15: Comparison between the volume-averaged source term $\tilde{\omega}_Y$ and the product of the flame surface and the (local) mass burning rate $\langle m \rangle \Sigma$ for the premixed flame (upper left) and stratified flames. $\tilde{\omega}_Y$ and $\langle m \rangle \Sigma$ are normalized with the mean $\tilde{\omega}_Y$ of all 15 realizations.

PDF shapes to model $\langle m \rangle$:

$$\langle m \rangle_\beta = \int_0^1 m(\phi(Z)) \mathcal{P}_\beta \left( Z; \tilde{Z}, \tilde{\omega}_Y \right) \, dZ,$$

$$\langle m \rangle_\delta = \int_0^1 m(\phi(Z)) \mathcal{P}_\delta \left( Z; \tilde{Z} \right) \, dZ,$$  

(3.27)  

(3.28)
In equations (3.27-3.28) $Z$ is normalized to enable the use of the (bounded) $\beta$-PDF, e.g., for cases TSI and TSII $Z=0$ corresponds to $\phi = 0.4$ and $Z=1$ corresponds to $\phi = 1.0$ while for case TSIII $Z=0$ corresponds to $\phi = 0.1$ and $Z=1$ corresponds to $\phi = 1.3$. Subfilter variances become increasingly important with increasing filter width; they are taken into account by equation (3.27) but not by equation (3.28). In LES, typically a transport equation for the FA mixture fraction $\overline{Z^n}$ is solved, while for the FA mixture fraction variance $\overline{Z^{\alpha n}}$ one can choose between a transport equation or an algebraic closure. The use of a similarity or gradient model, similar to the gradient model for the turbulent stresses, is common to estimate $\overline{Z^{\alpha n}}$:

\[
\overline{Z^{\alpha n}} = \alpha \Delta x^2 \left( \frac{\partial \overline{Z}}{\partial x_j} \right)^2 ,
\]

(3.29)

where $\alpha$ is often assumed to be a constant value. For an equidistant grid spacing and smooth scalar fields a Taylor-expansion yields $\alpha = 1/12$ but previous DNS results for turbulent subfilter stress models [Clark1979] or subfilter scalar variances [Branley2001] pointed out that this value underestimates variances. In other studies constant values for $\alpha$ ranging from $\alpha = 0.1$ [Branley2001] to $\alpha = 0.3$ [Vreman2008] are reported. To ensure accurate estimations of $\overline{Z^{\alpha n}}$, the appropriate value for $\alpha$ was determined by comparison of $\overline{Z^{\alpha n}}$-values extracted from DNS data and predictions by equation (3.29) in the same DNS scalar fields. For $\Delta_f$ equal to $\delta_0$, $2\delta_0$, $4\delta_0$ and $8\delta_0$ respective values for $\alpha$ equal to 0.17, 0.19, 0.23 and 0.58 are obtained. For premixed Bunsen flames, Vreman et al. [Vreman2008] obtained $\alpha = 0.12$ for $\Delta_f = 0.7 \delta_0$ and $\alpha = 0.17$ for $\Delta_f = 1.4 \delta_0$ using equation (3.29) to estimate $\overline{Z^{\alpha n}}$. These values comply well with values obtained in this study. Figure 3.16 depicts subfilter PDF’s of $\phi$ for two arbitrarily chosen filter volumes. In the left graph it can be seen that the estimated value for $\overline{Z^{\alpha n}}$ closely resembles the value extracted from the DNS. For the the filter volume shown on the right, a slight discrepancy is observed: the underestimated value for $\overline{Z^{\alpha n}}$ leads to a more contracted shape of the $\beta$-PDF. Nevertheless, a very good correspondence between the mean value for $\overline{Z^{\alpha n}}$ obtained from filtered DNS data and the mean predicted values. Equation (3.29) will therefore be used in the remainder of this study to estimate $\overline{Z^{\alpha n}}$ with the values for $\alpha$ extracted from DNS data. In figure 3.17 the conditional filtered FSD source term $\left( \langle m \rangle \right) \left[ Y_{eq} \left( Z \right) - Y_0 \left( Z \right) \right] \Sigma \delta_y \Sigma x$ is compared to $\overline{Z \delta_y}$ for a $\Delta_f$ equal to $\delta_0$ and $8\delta_0$. Filtered numerical data from the DNS are

**Figure 3.16:** Probability density function profiles for $\phi$ in two different filter volumes, both having $\Delta_f = 8 \delta_0$, for flame TSIII ($0.1 \leq \phi \leq 1.3$). Solid lines denote PDF’s extracted from DNS, dashed lines denote $\beta$-PDF’s using means and variances extracted from DNS and open symbols denote $\beta$-PDF’s using means from DNS and estimated variances using equation 3.29.
clustered in 30 bins in $\overline{\omega}_y$-direction; bins containing four data points or less are discarded. From the error bars it is clearly visible that the FSD-method described by equation (3.26)
yields a contribution close to $\overline{\omega Y}$ not just filtered over the entire computational domain, as shown in section 3.5.1, but on smaller filter volumes as well.

For case TSII, which exhibits the most intense mixing behavior as pointed out in section 3.4.4, a subfilter $\delta$-PDF yields almost identical results as a subfilter $\beta$-PDF with an estimated $Z^{\eta_2}$, as can be seen in figure 3.18. Both the profile for the FSD source term using the $\beta$- and $\delta$ PDF in figure 3.18 correspond very well with $\overline{\omega Y}$ extracted from the DNS data, depicted in figure 3.17. The curved shape of the profile originates from the fact that for filter volumes having a small value for $\overline{\omega Y}$ no flame surface is found. The entire chemical source is contracted in the flame sheet around $C \approx 0.7$ leading to an overestimation of the chemical source term by the FSD-model at higher values of $\overline{\omega Y}$. The difference between $\langle m \rangle [\gamma_{eq}(Z) - \gamma_0(Z)] \Sigma$ and $\overline{\omega Y}$ is more apparent for small filter widths where this effect is prevailing.

For flame TSIII, having the largest stratification ($0.1 \leq \phi \leq 1.3$), and to a lesser extend for flame TSI, the replacement of $\langle m \rangle$ by $\langle m \rangle_\delta$ results in an overestimation of the FSD source term when large filter widths are used. The use of a subfilter $\beta$-PDF for $m$, i.e. replacement of $\langle m \rangle$ by $\langle m \rangle_\beta$, still closely reproduces $\overline{\omega Y}$. The explanation lies in the nonlinear relation between $m$ and $\phi(Z)$ and the observation that subfilter variances become non-negligible for large filter widths. For large filter widths, implying relatively high subfilter variances, a $\delta$-peak will be put at intermediate $Z$-values where $m$ obtains high values. Thereby regions where $m$ is small are totally neglected and thereby this results in a significant overestimation of the FSD source term. This overestimation is clearly visible in figure 3.18.

It is concluded that the FSD model using a $\beta$-PDF constructed from FA mixture fraction moments, i.e. $\overline{Z}$ and $Z^{\eta_2}$, yields very good predictions for the filtered mass burning rate in the simulated stratified flames. Due to the nonlinear relation between $m$ and $\phi(Z)$, the replacement of the $\beta$-PDF by a $\delta$-PDF results in an overestimation of the FSD chemical source term when subfilter variances are non-negligible. This effect is less striking for flame TSI (not shown) than for flame TSIII since in flame TSI ($0.4 \leq \phi \leq 1.0$) the non-linearity in $m$ is smaller than in flame TSIII ($0.1 \leq \phi \leq 1.3$).

3.5.3 Application of a simple algebraic model for $\Sigma$

The final constituent of the FSD-model is a model for $\Sigma$. As mentioned in section 3.1, several model types for $\Sigma$ have been proposed for LES purposes. The class of simple algebraic models for $\Sigma$ can generally be written in a generic form:

$$\Sigma = \sigma \overline{\ell} \left( 1 - \overline{\ell} \right),$$  \hspace{1cm} (3.30)

in which $\sigma$ is a function of properties from the turbulent flame brush. Here, $\sigma$ is assumed to be constant and is extracted from the scatter plots in figure 3.14 using a method of least squares. For flames TP, TSI, TSI and TSIII values for $\sigma$ equal to 4585, 4935, 4754 and 5302 [m$^{-1}$] are obtained, respectively. The dashed lines in the scatter plots in figure 3.14 represent the function given by equation (3.30) using the applicable value for $\sigma$. Figure 3.19 compares the FSD-model given by equation (3.27) using $\Sigma$ extracted from the DNS data and $\Sigma$ modeled by equation (3.30). It is observed that the algebraic model for $\Sigma$ overestimates small values for the chemical source term while large values are underestimated. Overestimation of small values for the chemical source term occurs for $0.9 \leq \overline{\ell} \leq 1.0$: when values $\overline{\ell} \geq 0.9$ are neglected, overestimation of small values of the
chemical source term is significantly reduced for all flames. The systematic underestimation of large values for the chemical source term comes from the fact that the maximum chemical source term obtained by equation (3.30) equals $m\sigma/4$ where $m$ denotes the maximum mass burning rate, occurring at $\phi = 0.911$. Every deviation from these values will result in a decrease of the chemical source term since both $m$ and $\Sigma$ exhibit a quasi-parabolic behavior around this maximum. For flame TP $m = m_0$ since only $\phi = 0.7$ occurs.

In figure 3.19 can be seen that using the FSD approach, in which $\Sigma$ is modeled by equation (3.30) and the filtered mass burning rate is modeled by $\langle m \rangle_\beta$, a good correspondence with $\bar{\omega}_Y$ is obtained. Besides the minor overestimation of $\Sigma$ for $0.9 < \tilde{C} \leq 1.0$ this FSD approach only deviates from $\bar{\omega}_Y$ for high values of $\bar{\omega}_Y$. The theoretical upper limit of the $\Sigma$-value dictated by equation (3.30) is the cause of this underestimation.

### 3.6 Conclusions

In this study the applicability of the Flame Surface Density (FSD) approach, developed for premixed flames, to flames with an equivalence ratio variation has been analyzed. Three stratification setups were used in which the mixing length scale was altered by decreasing the stratification length scale or increasing the range of $\phi$. The extension of the range of $\phi$, besides the reduced mixing length scale, also leads to a larger variation in the mass burning rate $m$. Direct Numerical Simulations (DNS) of the turbulent stratified Bunsen flames were enabled by the use of the Flamelet Generated Manifold (FGM) reduction method for chemical kinetics.

All turbulent flames, having $35.1 \leq Re_{\lambda} \leq 37.4$ and $33.8 \leq Ka \leq 37.7$, are in the Thin
Reaction Zones (TRZ) regime and flame structures are nearly identical to premixed flamelets. Flame curvature has a mean length scale approximately one order of magnitude larger than the flame thickness. Therefore it is assumed to be justified to use stretchless flamelets for combustion chemistry representation. The applied stratifications did not cause any deviations from premixed flamelet profiles either: mean mixing length scales were approximately one order of magnitude larger than the flamelet flame thickness for ($\phi = 0.7$) implying a premixed combustion mode.

The FSD-model proved to be applicable to stratified flames when the variation in mass burning rate with $\phi$ is taken into account. Considering the entire computational domain, the FSD-model obtained values for the chemical source term having a very similar accuracy for the premixed and stratified flames. If the subfilter variation of $m$ is subsequently modeled by a $\beta$-PDF, i.e. $\langle m \rangle = \langle m \rangle_{\beta}$, a good correspondence between the filtered progress variable source term and the FSD-model was obtained. For small filter widths the clustering of the entire chemical source in an infinitely thin flame sheet leads to an underestimation of small chemical source terms and an overestimation of large chemical source terms. These effects become less prevalent when large filter widths are considered. When $\langle m \rangle_{\beta}$ is replaced by $\langle m \rangle_{\delta}$ only flame TSIII shows a significant deviation when large filter widths are considered. Large filter widths imply a large subfilter mass burning rate variation and an intermediate value for the $\tilde{Z}$. The latter, in conjunction with the $\phi$-$m$ profile shown in figure 3.13, results in a systematic overestimation of the filtered mass burning rate.

When for the flame surface density $\Sigma$ a simple algebraic Bray-Moss-Libby (BML) model is employed, only two systematic alterations of results are observed. An overestimation of $\Sigma$ for $0.9 \leq \tilde{\zeta} \leq 1.0$ leads to a small, but perceptible, overestimation of small chemical source terms. High values of the chemical source term are underestimated due to the limited maximum $\Sigma$-value dictated by equation (3.30). Nevertheless the FSD-model, using a $\beta$-PDF to account for the subfilter mass burning rate variation and a simple algebraic model for $\Sigma$, already yields a good agreement with the filtered source term obtained from filtered DNS data. Results are assumed to get even better when a more sophisticated model for $\Sigma$ is used.
A priori analysis of FGM tables

All flames studied in chapter 3 remained fully within flammability limits. Based on this fact and results from previous studies [Bongers2005-2, Fiorina2005-1], the choice to use premixed flamelets to construct the FGM table was an obvious one. In this chapter partially-premixed flames of which the fuel and oxidizer are outside flammability limits are treated. The question which flamelet type is most appropriate for modeling these flames, with a significant larger range in $\phi$, is addressed in this chapter.

The objective of this chapter is to assess the accuracy of representation of chemical kinetics by low-dimensional FGM databases for turbulent partially-premixed jet flames. In order to eliminate errors originating from CFD simulations, an a priori analysis of FGM databases is performed in which FGM databases are directly compared to experimental results. It is investigated to what extent the used flamelet structure type (premixed or counterflow diffusion flamelets) and the molecular transport model (multi-component diffusion or unit Lewis numbers) should resemble the characteristics of the analyzed turbulent flame to yield an accurate representation of combustion chemistry. The influence of the flamelet type and the molecular transport model on the accuracy of mass fraction predictions will be assessed for Sandia Flames C, D, E and F [Barlow2003], which exhibit behavior ranging from moderately turbulent (Flame C, $Re = 13.400$) to heavily turbulent with local extinction and re-ignition (Flame F, $Re = 44.800$). More specifically, the focus will be on predictions of CO, H$_2$ and OH since these are still hard to predict accurately.

4.1 Introduction

For FGM databases often the assumption is made that the flame structure, on which the FGM is based, is either premixed or non-premixed. Partially-premixed flames, which exhibit features from both premixed and non-premixed structures, it is not known beforehand whether premixed or non-premixed flame structures are more appropriate to model the flame.

Previously Chen et al. [Chen1992] showed that a chemistry database, generated from perfectly stirred reactor computations, overestimated CO mass fractions in rich parts of a methane/air diffusion flame. Pitsch and Steiner [Pitsch2000] performed LES simulations of Sandia Flame D using a Lagrangian Flamelet Model (LFM) incorporating diffusion in mixture fraction direction. A database was generated and parameterized on mixture fraction ($Z$) and scalar dissipation rate ($\chi$). To incorporate subfilter influences on chemical
A priori analysis of FGM tables

kinetics a Presumed PDF method using a marginal \( \beta \)-PDF for \( Z \) is used. \( \chi \) is subsequently computed from the \( Z \) field. Conditional mean \( \text{CO}_2, \text{CO}, \text{H}_2\text{O}, \text{H}_2 \) and \( \text{OH} \) mass fractions were presented of which \( \text{CO}_2 \) was very well predicted. The conditional mean \( \text{H}_2\text{O} \) mass fraction was underpredicted for \( Z \) values larger than the stoichiometric value while both \( \text{CO} \) and \( \text{H}_2 \) were overestimated under fuel-rich conditions. Especially at \( x = 15D \) the maximum \( \text{OH} \) mass fraction was significantly overestimated. It was argued that premixing of the fuel with air occurred to fast causing combustion reactions to take place too early. This, in turn, led to a slight overprediction in formation of \( \text{H}_2\text{O} \) and the intermediates \( \text{H}_2 \) and \( \text{CO} \) in the fuel-rich region. The overestimation of mixing in the flow field is here said to cause the discrepancy in species mass fractions predictions. Dally et al. [Dally2003] confirmed that predictions of \( \text{CO} \) in fuel-rich conditions are difficult. Vervisch et al. [Vervisch2004] simulated Sandia Flame D using a combination of FPI-/diffusion flamelet approach. During the simulation chemical source terms were retrieved from the FPI-database (based on premixed flamelets) which was parameterized on \( Z \) and a reaction progress variable (\( Y \)). Species mass fractions were interpolated a posteriori from the diffusion flamelet database. To model the influence of subfilter fluctuations of combustion chemistry a Presumed PDF (PPDF) closure method was adopted using a marginal \( \beta \)-PDF for both \( Z \) and \( Y \). Only species mass fractions results for \( \text{CO} \) were shown in mixture fraction space for Sandia Flame D at heights \( x = 3.0D, x = 15D \) and \( x = 60D \) from the inlet plane. At \( x = 15D \), which is by far the most interesting because information is available for the entire range in mixture fraction, conditional mean \( \text{CO} \) mass fractions were slightly overestimated on the fuel-rich side and a significant underestimation occurred at \( Z \approx 0.4 \). The same deviations from measurements were observed in conditional mean \( Y \) values; these deviations were attributed to various factors of which one was the chemical scheme in combination with the used (premixed flamelet based) FPI tabulation method.

Bongers et al. [Bongers2005-1] showed that a FGM database based on premixed flamelets can accurately describe the premixed part of partially-premixed counterflow flames. However, in this study the considered range in mixture fraction was limited since the fuel stream had the same composition as a premixed system at the upper flammability limit. Fiorina et al. [Fiorina2005-1] analyzed how well a FPI database, also parameterized on \( Z \) and \( Y \), represented complex chemistry for methane/air premixed, partially-premixed and diffusion flames. For the premixed flame it was found that the FPI database reproduced results from detailed chemistry very well. However, for the partially-premixed and diffusion flames significant differences for temperature, \( \text{CO}_2 \) and \( \text{CO} \) mass fractions were observed between the FPI predictions and detailed chemistry for fuel-rich conditions. Apparently a premixed flamelet-based database cannot represent chemical kinetics accurately for partially-premixed and diffusion flames in fuel-rich conditions. Vreman et al. [Vreman2008] performed LES simulations of Sandia Flames D and F using a premixed flamelet-based and a counterflow diffusion flamelet-based FGM database. It was found that \( \text{CO}_2, \text{H}_2\text{O}, \text{CH}_4 \) and \( \text{O}_2 \) mass fractions were well-predicted but \( \text{CO}, \text{H}_2 \) and \( \text{OH} \) mass fraction were predicted less satisfactory although the counterflow diffusion flamelet-based FGM yielded significantly better results than the premixed flamelet-based FGM. This study confirms observations from Bradley et al. [Bradley2002], who also obtained a significant overestimation of \( \text{CO} \) and \( \text{H}_2 \) mass fractions when using a premixed flamelet library. Results by Vreman et al. using the counterflow diffusion flamelet-based FGM were in good accordance with outcomes obtained by Sheikh et al. [Sheikh2005] who also used non-premixed flamelets to construct a thermochemical table. Although Vreman et al. [Vreman2008] found that the inclusion of modeled subfilter variances of mixture
fraction and progress variable as additional entries to the manifold had only small effects on the simulation of both Sandia Flame D and E, the question remains whether the error in CO, H2 and OH mass fractions originates from the tabulated chemistry or from the LES simulation.

In LES or RANS simulations of turbulent reacting flow using tabulated chemistry combined with a subfilter model, three main origins of errors can be distinguished. The first origin of errors can be attributed to an inappropriate representation of chemical kinetics by the tabulated chemistry, subsequently the subfilter modeling assumptions used in transport equations can be the origin of inaccuracies in numerical simulations. The third class of errors are made during the actual CFD simulation: the accuracy of the used numerical schemes and convergence of the results.

Chapter outline

In the next section the generation of the FGM databases will be discussed for both premixed- and counterflow diffusion flamelet-based databases. This is followed by the analysis of the generated FGM databases which is split up into three parts: in the first part a direct comparison between measured and predicted species mass fractions and in the second part an analysis conditioned on Z to gain more insight on the underlying physics. In the third part an analysis of which type of FGM database can adequately describe differential diffusion effects will be performed; this analysis is performed to determine the appropriate molecular transport model to be used in FGM databases for current test cases. The paper ends with a recommendation which type of FGM database should be used to simulate the Sandia Flames and the accuracy of CO, H2 and OH mass fraction predictions when using the best-suited FGM database.

4.2 Construction of FGM databases

Before FGM databases can be compared with experimental data they first have to be generated. In this section the equations that describe the flamelet solutions of which FGM’s are composed are dealt with. Boundary conditions for the different types of flamelets to be used and methods to construct FGM’s from multiple flamelet solutions are subsequently discussed.

For this study, partially-premixed combustion parameters are mapped on two control variables describing mixing (mixture fraction Z) and reaction progress (reaction progress variable Y), i.e.

\[ \varphi = \varphi (Z, Y) \]  

in which \( \varphi \) can denote any thermochemical variable. The mixture fraction \( Z \) is defined by Bilger [Bilger1990] which was modified by Barlow et al. [Barlow2005] by removing the oxygen terms and thereby making the mixture fraction less sensible to experimental noise and interference from laser-induced fluorescence [Barlow1998]. The proposed, modified mixture fraction definition reads:

\[ Z = \frac{2 M_H^{-1} [Z_H - Z_{H,2}] + 0.5 M_C^{-1} [Z_C - Z_{C,2}]}{2 M_H^{-1} [Z_{H,1} - Z_H] + 0.5 M_C^{-1} [Z_{C,1} - Z_C]} \]  

(4.2)
in which \( M_j \) denotes the molar mass of an element \( j \), \( Z_j \) denotes the mass fraction of an element and subscripts \( H, C, 1 \) and \( 2 \) refer to hydrogen, carbon and the fuel- and oxidizer stream, respectively. The reaction progress variable, which has to be monotonous in both lean and rich mixtures in order to facilitate an unambiguous determination of dependent variables, for the Sandia Flames is defined as:

\[
Y = \sum_{i=1}^{N_s} \alpha_i Y_i , \tag{4.3}
\]

in which \( \alpha_i = 0 \) for all species except for \( \text{CO}_2 \), \( \text{H}_2\text{O} \) and \( \text{H}_2 \). Weight factors for these species are \( \alpha_{\text{CO}_2} = M^{-1}\text{CO}_2 \), \( \alpha_{\text{H}_2\text{O}} = M^{-1}\text{H}_2\text{O} \) and \( \alpha_{\text{H}_2} = M^{-1}\text{H}_2 \). Values for \( \alpha_i \) are not trivial; it was found that presented weight factors ensured a monotonous increasing \( Y \) for all flamelets used.

The equations describing steady, stretchless premixed flamelets have been derived in section 2.2.1 and read:

\[
\frac{\partial \rho u}{\partial x} = 0 , \tag{4.4}
\]
\[
\frac{\partial \rho u Y_i}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda}{L e_i c_p} \frac{\partial Y_i}{\partial x} \right) + \dot{\omega}_i , \quad i \in [1, N_s - 1] \tag{4.5}
\]
\[
\frac{\partial \rho u h}{\partial x} = \frac{\partial}{\partial x} \left[ \frac{\lambda}{c_p} \frac{\partial h}{\partial x} + \sum_{i=1}^{N_s} \left( \frac{1}{L e_i} - 1 \right) h_i \frac{\partial Y_i}{\partial x} \right] , \tag{4.6}
\]
\[
\rho = \rho_{\infty} M / (R T) , \tag{4.7}
\]

in which the Low Mach-number approximation is used in the thermal equation of state (4.7) with \( \rho_{\infty} \) denoting the ambient pressure. Dirichlet boundary conditions for \( Y_i \) and \( h \) are imposed at the unburnt side:

\[
Y_i (x \to -\infty) = Y_{i,-\infty} , \quad h (x \to -\infty) = h_{-\infty} , \tag{4.8}
\]

and Neumann boundary conditions for \( Y_i \) and \( h \) are imposed at the chemical equilibrium side:

\[
\frac{\partial Y_i}{\partial x} (x \to \infty) = 0 , \quad \frac{\partial h}{\partial x} (x \to \infty) = 0 . \tag{4.9}
\]

The translational degree of freedom is removed by choosing a fixed temperature at \( x = 0 \). Each flamelet contained 200 points and the premixed flamelet based FGM databases contains 450 flamelets between the numerical flammability limits; for the Sandia Flames the lower numerical flammability limit was found at \( Z = 0.1 \) and the upper flammability limit was still not reached at \( Z = 1 \). Between the lower flammability limit, thermochemical variables are linearly interpolated between the leanest flamelet and pure oxidizer. Chemical equilibrium has been calculated and explicitly added to all flamelets to ensure equilibrium mass fractions for CO and \( \text{CO}_2 \). It must be noted that when multi-component diffusion is employed, \( Z \) is not constant in a premixed flamelet due to differential diffusion effects.

The equations describing quasi-steady, counterflow diffusion flamelets were derived in
4.2 Construction of FGM databases

\[ \frac{\partial \rho}{\partial t} + \frac{\partial \rho u}{\partial x} = -\rho G , \] (4.10)
\[ \frac{\partial \rho Y_i}{\partial t} + \frac{\partial \rho u Y_i}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda}{L_{e_i} c_p} \frac{\partial Y_i}{\partial x} \right) + \dot{\omega_i} - \rho Y_i G, \quad i \in [1, N_s - 1] , \] (4.11)
\[ \frac{\partial \rho h}{\partial t} + \frac{\partial \rho u h}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda}{c_p} \frac{\partial h}{\partial x} + \frac{\lambda}{c_p} \sum_{i=1}^{N_s} \left( \frac{1}{L_{e_i}} - 1 \right) h_i \frac{\partial Y_i}{\partial x} \right) - \rho h G , \] (4.12)
\[ \rho \frac{\partial G}{\partial t} + \rho u \frac{\partial G}{\partial s} = \frac{\partial}{\partial s} \left( \mu \frac{\partial G}{\partial s} \right) + \rho_{\text{ox}} \left( \frac{\partial a(t)}{\partial t} + a(t)^2 \right) - \rho G^2 . \] (4.13)

in which strain rate \( G \) is defined as \( G \equiv \frac{\partial \mathbf{v}}{\partial y} \). The derivation of equation (4.13) can be found in appendix A. The Low Mach-number approximation for the equation of state, equation (4.7), is used to close the set of equations. Dirichlet boundary conditions for \( Y_i, h \) and \( G \) are imposed in the oxidizer \((x \to -\infty)\) stream while in the fuel \((x \to \infty)\) stream a Neumann boundary condition for \( G \) is imposed:

\[ Y_i(x \to \infty) = Y_{i,\text{ox}} , \quad h(x \to \infty) = h_{\text{ox}} , \quad G(x \to \infty) = a(t) . \] (4.14)
\[ Y_i(x \to -\infty) = Y_{i,\text{fu}} , \quad h(x \to -\infty) = h_{\text{fu}} , \quad \frac{\partial G}{\partial x}(x \to -\infty) = 0 . \] (4.15)

The parameter of this system is the applied strain rate \( a(t) \), which is defined at the oxidizer side \((x = \infty)\). The translational degree of freedom is removed by choosing the stagnation plane at \( x = 0 \). Please note that counterflow diffusion flamelets are computed in physical space instead of \( Z \)-space. This has the advantage that no assumption has to be made for the profile of the scalar dissipation rate \( \chi \), which occurs when equations (4.11-4.12) are transformed to \( Z \)-space.

Because counterflow diffusion flamelets extinguish at a limiting strain rate, the region in \( Z-Y \) space between the maximum strain rate and the mixing limit cannot be spanned by steady counterflow diffusion flamelets. The extinction strain rate equals \( a = 657 \) [1/s] for a multi-component diffusion transport model and \( a = 978 \) [1/s] for a unit Lewis number transport model; this limit is shown as the lower blue line in figure 4.2. To guarantee a smooth continuation at the extinction limit, the applied strain rate is a function of time, e.g. \( a = a(t) \) with a small value \((a = 2 \) [1/s]) as initial value. The strain rate is subsequently increased in time using an exponential function with a timescale that is significantly larger than that of the slowest evolving species: it is verified that quasi-steady counterflow diffusion flamelet solutions having a strain rate below the extinction strain rate are (nearly) identical to their steady flamelet solutions. Time-dependent solution of equations (4.10-4.13) is tracked until the flame is extinguished and the solution equals the mixing solution. For a quenching counterflow diffusion flamelet \( \dot{Y} \) decreases: the use of these unsteady flamelets does not require an additional control variable.

Both the premixed and counterflow diffusion flamelets are interpolated onto an identical \( Z-Y \) grid: in this study a \( 201 \times 201 \) \((Z \times Y)\) equidistant discretization is used and data is retrieved using linear interpolation in both directions. As an example water (H\(_2\)O) and hydroxyl (OH) from the premixed flamelet database, using unit Lewis numbers, are shown in figure 4.1. For all flamelets, detailed chemistry is modeled by the GRI 3.0 reaction mechanism [GRI mech30] and fuel and oxidizer compositions have been taken from Liu et al. [Liu2005].
4.3 A priori validation of FGM databases

The availability of extensive experimental data on Sandia Flames C, D, E and F by Barlow et al. [Barlow2003] allows a priori validation of FGM databases. The experimental data consists of several thousands of instantaneous measurements of 9 species (O$_2$, N$_2$, H$_2$, H$_2$O, CH$_4$, CO$_2$, CO, H$_2$ and NO) at different heights and radial positions in all four flames. For every instantaneous measurement $Z$ and $Y$ are computed according to equation (4.2) and (4.3); resulting scatter plots are shown together with the boundaries of the premixed flamelet-based FGM database (red line) and counterflow diffusion flamelet-based FGM database (blue lines) in figure 4.2 for Sandia Flame C and F. Using these reconstructed values for $Z$ and $Y$, corresponding thermochemical data can be retrieved from the FGM database. Subsequently experimentally observed averaged species mass fractions, indicated by $Y_i^{\text{EXP}}$, can be compared to averaged species mass fractions retrieved from the FGM database which are indicated by $Y_i^{\text{FGM}}$. It must be remarked that only measurement data from nine species, O$_2$, CH$_4$, N$_2$, H$_2$O, CO$_2$, H$_2$, CO, OH and NO, is available with measurement uncertainties equal to 0.4%, 0.5%, 3%, 4%, 4%, 10%, 10%, 10% and 15% respectively. The sum of all measured mass fractions is less than 0.2% away from unity for all instantaneous measurements at all probed locations, implying that mass fractions of species which have not been measured do not have a significant influence on the reconstructed value for $Z$ and $Y$.

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4.3 A priori validation of FGM databases

When data is retrieved from the FGM databases the experimentally observed \( Z \) is bounded between zero and unity and \( Y \) is bounded to its minimum and maximum values as found in the FGM database. In figure 4.2 it can be seen that around \( Z = 0.4 \) a significant number of super-equilibrium points is observed. In this analysis these super-equilibrium measurements are replaced by corresponding maximum \( Y \) values as found in the FGM database.

To allow the validation of a laminar combustion chemistry database the spatial resolution of measurements must be significantly smaller than the chemical length scale present in the turbulent flame. Spatial resolution of measurements has been estimated to be approximately 750 \( \mu m \) [Barlow1998]. A quick estimation of the chemical length scale tells us that it is of the same order of magnitude as the probe resolution:

\[
L_{\text{chem}} \propto \sqrt{\frac{D}{a}} = \sqrt{\frac{O(10^{-5})}{O(10^2)}} = O(10^{-3})[m] \sim L_{\text{probe}}
\]

(4.16)

In the discussion at the end of Barlow et al. [Barlow1998] it is argued that spatial averaging over the probe volume has very limited influence on measured scalar quantities: spatial averaging effects are only expected to have a non-negligible influence in high-gradient regions close to the inflow plane (\( z \leq 3D \)). For this study it is therefore assumed that spatial averaging over the probe volume is of sufficiently small influence to be neglected.

In this section results of a priori analysis of FGM databases for predictions of \( \text{H}_2\text{O}, \text{H}_2, \text{CO}_2, \text{CO} \) and \( \text{OH} \) will be discussed of which \( \text{H}_2, \text{CO} \) and \( \text{OH} \) will be treated more in-depth. Four different FGM databases will be compared as stated in table 4.1. Scatter plots will be presented to gain a global insight on the performance of different types of FGM databases; subsequently conditional (on mixture fraction) plots will give more insight to explain underlying physical phenomena.

### 4.3.1 Direct comparison of measured with predicted mass fractions

Direct comparison of experimentally observed with predicted mass fraction yield a quick, but not fully detailed, insight on the predictive accuracy of a FGM database. To allow quantification of the accuracy a definition for the error is introduced:

\[
\varepsilon_i = \sqrt{\frac{1}{N} \sum_{k=1}^{N} (Y_{i}^{\text{EXP}} - Y_{i}^{\text{FGM}})^2 \left[ \frac{1}{N} \sum_{k=1}^{N} Y_{i}^{\text{EXP}} \right]^{-1}}
\]

(4.17)

which equals the RMS-value of a species \( i \) its mass fraction normalized by its average experimentally observed mass fraction. The definition contains a summation over

<table>
<thead>
<tr>
<th>Designation</th>
<th>Flamelet type</th>
<th>Molecular transport</th>
</tr>
</thead>
<tbody>
<tr>
<td>FGM-PL</td>
<td>Premixed</td>
<td>Unit Lewis numbers</td>
</tr>
<tr>
<td>FGM-PC</td>
<td>Premixed</td>
<td>Multi-component diffusion</td>
</tr>
<tr>
<td>FGM-DL</td>
<td>Counterflow diffusion</td>
<td>Unit Lewis numbers</td>
</tr>
<tr>
<td>FGM-DC</td>
<td>Counterflow diffusion</td>
<td>Multi-component diffusion</td>
</tr>
</tbody>
</table>

**Table 4.1:** Designations for the four different FGM databases used in this study.
A priori analysis of FGM tables

instantaneous measurements; here the summation has been taken over all heights and radial positions for a specific flame. Differences between measured and predicted values can be caused by either measurement uncertainties and/or discrepancies between the experimentally observed- and tabulated chemistry. Measurement uncertainties cause both a spreading in measurement data and, subsequently, an inaccuracy for \( Z \) and \( \gamma \) which are reconstructed according to equation (4.2) and (4.3). An inaccuracy in reconstructed \( Z \) and \( \gamma \) implies that data is retrieved from a not-exactly-right location in the FGM database. Discrepancies between the experimentally observed- and tabulated chemistry introduces a third origin for differences between instantaneous measurements and corresponding data from FGM databases. In order to minimize differences between (instantaneous) measurements and predictions, measurement uncertainties need to minimized and the type of FGM database should be tuned to the type of flame which is to be simulated. For averaged values the dispersion of instantaneous measurements introduces an additional error when tabulated thermochemical variables exhibit a curved profile in \( Z-\gamma \) space. This error can be estimated using a Taylor expansion:

\[
\delta Y_i \approx \frac{\partial Y_i}{\partial Z} \delta Z + \frac{\partial Y_i}{\partial \gamma} \delta \gamma + \frac{1}{2} \left[ \frac{\partial^2 Y_i}{\partial Z^2} (\delta Z)^2 + \frac{\partial^2 Y_i}{\partial \gamma^2} (\delta \gamma)^2 + \frac{\partial^2 Y_i}{\partial Z \partial \gamma} \delta Z \delta \gamma \right]
\]  

(4.18)

From equation (4.18) can be seen that when a thermochemical variable is sufficiently smooth, i.e. having small higher order terms, an error in individual \( Z \) and/or \( \gamma \) measurements does not cause a significant error in the averaged value of the thermochemical variable since positive and negative errors cancel out if \( \delta Z \) and \( \delta \gamma \) are symmetrically distributed. When the higher order terms in equation (4.18) are not negligible, uncertainties in individual \( Z \) and/or \( \gamma \) measurements will cause a non-negligible error in the averaged value of retrieved variables. In figure 4.1, \( H_2O \) and \( OH \) are shown in \( Z-\gamma \) space: \( H_2O \) has negligible higher order terms while for \( OH \) these terms are far from negligible and will likely cause an underestimation of maximum \( OH \) mass fractions and an overestimation of small \( OH \) mass fractions.

In order to determine systematic errors, conditionally (ensemble) averaged predicted mass fractions are presented together with standard deviations of instantaneous measurements. To determine these averaged mass fractions measured mass fractions are clustered in 40 equidistant bins. Bins containing less than 25 data points are not considered in this analysis due to the lack of statistical convergence of these bins. For each instantaneous measurement, corresponding data is retrieved from FGM databases as a function of \( Z \) and \( \gamma \) which are reconstructed from measured mass fractions. Subsequently the data retrieved from FGM databases is averaged bin-wise. This data is then compared to experimental data which is also averaged with the same procedure.

\( H_2O \) mass fractions can be predicted very accurately using any type of FGM. To illustrate this, the test cases with the minimum and maximum error, as defined in equation (4.17), are shown in figure 4.3. The well predicted averaged value and the negligible standard deviation can be attributed to the small measurement uncertainty for \( H_2O \) and the smooth behavior of \( H_2O \) in \( Z-\gamma \) space. The only deviation from the \( Y_i^{FGM} = Y_i^{EXP} \) line can be seen for the maximum measured mass fractions which do not occur in any FGM database. It can be concluded that predictions for \( H_2O \) are rather insensitive to the type of FGM which is used.

For \( H_2 \) mass fractions, shown in figure 4.4, the premixed flamelet-based FGM databases, and especially the FGM-PC, yield a significant overestimation. Standard deviations are
4.3 A priori validation of FGM databases

Figure 4.3: Conditional averages (circles) together with standard deviations (error bars) of predicted $\text{H}_2\text{O}$ mass fractions (vertical axis) versus measured mass fractions. The horizontal dashed red line indicates the maximum $\text{H}_2\text{O}$ mass fraction present in the FGM database. The situation with the smallest error is shown on the left, the situation with the largest error is shown on the right.

Substantial and the averaged predicted $\text{H}_2$ mass fractions significantly exceeds the measured mass fractions. Both counterflow diffusion flamelet-based FGM databases show smaller standard deviations and the predicted mass fractions correspond well to measured mass fractions. For large measured mass fractions ($Y_{\text{H}_2} > 0.004$) the FGM-DC tends to slightly underestimate $\text{H}_2$ mass fractions, this is not observed for the FGM-DL database. The reason the standard deviation is considerable is twofold: first of all the measurement uncertainty for hydrogen equals 10%, in very fuel-rich conditions this uncertainty can rise to 30% due to interference with hydrocarbons [Barlow2001]. Second of all the higher order derivatives of $\text{H}_2$ in $Z$-$Y$ space are not negligible resulting in errors in averaged values and standard deviations.

Scatter plots of measured $\text{CO}_2$ mass fractions versus predicted $\text{CO}_2$ mass fractions from premixed flamelet-based FGM databases show a two-branch structure, of which one branch shows a significant underestimation of $\text{CO}_2$ mass fractions. The branch exhibiting a significant underestimation of $\text{CO}_2$ mass fractions corresponds to fuel-rich combustion and will be discussed more in-depth in section 4.3.2. This two-branch structure also causes the conditionally averaged predicted values to be significantly smaller than measured ones. Counterflow diffusion flamelet-based FGM databases yield better predictions for $\text{CO}_2$ mass fractions than premixed flamelet-based FGM databases: standard deviations are substantially smaller. The FGM-DL database shows a better correspondence between the averaged mass fractions than the FGM-DC database: the latter shows a systematic underestimation of conditionally averaged $\text{CO}_2$ mass fractions. Measurement uncertainty for $\text{CO}_2$ is only 4% implying that errors mainly originate from reconstruction of $Z$ and $Y'$ and dissimilarities between the measured and tabulated chemistry.

From the averaged values in figure 4.6 can be seen that the FGM-PL database overestimates $\text{CO}$ mass fractions; this is also observed for other flames. The FGM-PC database exhibits the same trend but to a smaller extent. This overestimation of $\text{CO}$ mass fractions is to be expected because of the underestimation of $\text{CO}_2$ mass fractions while the element mass fraction of carbon must be conserved. Counterflow diffusion flamelet-based FGM databases yield more accurate predictions, although small $\text{CO}$ mass fractions
A priori analysis of FGM tables

Figure 4.4: Conditional averages (circles) together with standard deviations (error bars) of predicted H₂ mass fractions (vertical axis) versus measured mass fractions. The horizontal dashed red line indicates the maximum H₂ mass fraction present in the FGM database. The situation with the smallest error is shown on the left, the situation with the largest error is shown on the right.

<table>
<thead>
<tr>
<th>FGM type</th>
<th>(Y_{\text{H}_2})</th>
<th>(Y_{\text{H}_2})</th>
<th>(Y_{\text{CO}_2})</th>
<th>(Y_{\text{CO}})</th>
<th>(Y_{\text{OH}})</th>
</tr>
</thead>
<tbody>
<tr>
<td>FGM-PL</td>
<td>0.056</td>
<td>1.36</td>
<td>0.273</td>
<td>1.00</td>
<td>1.48</td>
</tr>
<tr>
<td>FGM-PC</td>
<td>0.089</td>
<td>2.38</td>
<td>0.343</td>
<td>0.769</td>
<td>1.34</td>
</tr>
<tr>
<td>FGM-DL</td>
<td>0.061</td>
<td>0.686</td>
<td>0.147</td>
<td>0.545</td>
<td>1.27</td>
</tr>
<tr>
<td>FGM-DC</td>
<td>0.070</td>
<td>0.833</td>
<td>0.237</td>
<td>0.784</td>
<td>1.41</td>
</tr>
</tbody>
</table>

Table 4.2: Maximum error as defined by equation (4.17) for different species and FGM databases.

\(Y_{\text{CO}} < 0.05\) tend to be slightly overestimated while large CO mass fractions \((0.05 < Y_{\text{CO}} < 0.085)\) tend to be underestimated. Substantial standard deviations are partly caused by the measurement uncertainty for CO which equals 10% and in very fuel-rich conditions can rise to 20%.

For both the premixed- and counterflow diffusion flamelet-based databases it was observed that a multi-component diffusion transport model yields reduced OH mass fractions when compared to databases with a unit Lewis number transport model. Since OH is assumed to be in quasi steady state the reduction of the maximum mass fraction, when a multi-component diffusion transport model is used instead of a unit Lewis number transport model, is most probably caused by differential diffusion of other species. For all FGM databases standard deviations are very large but for the FGM-PL, the FGM-PC and the FGM-DL databases can be seen that the conditionally averaged predicted OH mass fractions correspond well to the conditionally averaged measured OH mass fractions for \(Y_{\text{OH}} < 0.004\). For large OH mass fractions \((Y_{\text{OH}} > 0.004)\) the averaged mass fractions are underestimated; this can again be attributed to the fact that maximum measured mass fractions do not occur in the FGM database. The FGM-DC also exhibits this underestimation at large OH mass fractions, in addition small OH mass fractions \((Y_{\text{OH}} < 0.0025)\) are overestimated. The large standard deviations are caused by the uncertainty in experiments which serve as input, and a very localized OH peak in \(Z-Y\) space as can be seen in figure 4.1.

From the results above and the overview of errors in table 4.2 it can be concluded that
4.3 A priori validation of FGM databases

Figure 4.5: Conditional averages (circles) together with standard deviations (error bars) of predicted CO$_2$ mass fractions (vertical axis) versus measured mass fractions. The horizontal dashed red line indicates the maximum CO$_2$ mass fraction present in the FGM database. The situation with the smallest error is shown on the left, the situation with the largest error is shown on the right. Instantaneous measurements are shown to illustrate the two-branch structure of predictions from premixed flamelet-based FGM databases.

Figure 4.6: Conditional averages (circles) together with standard deviations (error bars) of predicted CO mass fractions (vertical axis) versus measured mass fractions. The horizontal dashed red line indicates the maximum CO mass fraction present in the FGM database. The situation with the smallest error is shown on the left, the situation with the largest error is shown on the right.

counterflow diffusion flamelet-based databases yield significant better predictions than premixed flamelet-based ones. Premixed flamelet-based databases tend to overestimate H$_2$ and CO mass fractions while underestimating CO$_2$ mass fractions. Comparison of results from the FGM-DL and the FGM-DC database shows that the FGM-DL yields more accurate results for the conditionally averaged species mass fractions. For OH mass fractions the FGM-PL, the FGM-PC and the FGM-DL database yield comparable results, both in conditionally averaged OH mass fractions and standard deviations. The FGM-DC database yields less accurate results for the conditionally averaged OH mass fractions than the other databases. For water the choice of FGM database has no significant influence on accuracy of predictions.
FIGURE 4.7: Conditional averages (circles) together with standard deviations (error bars) of predicted OH mass fractions (vertical axis) versus measured mass fractions. The horizontal dashed red line indicates the maximum OH mass fraction present in the FGM database.

4.3.2 Averaged mass fractions conditioned on $Z$

In the previous section predictive performance of the four different FGM databases, as stated in table 4.1, has been globally assessed for the Sandia Flames C to F. It was observed that for H$_2$, CO$_2$ and CO the FGM-PL and the FGM-PC database cannot represent observed mass fractions accurately. The counterflow diffusion flamelet-based FGM databases perform significantly better, especially the FGM-DL database. In this section predicted mass fractions will be compared to experimentally observed mass fractions for several heights in Flames C to F. Results will be presented conditioned on $Z$ in order to gain more insight in underlying physical phenomena. Experimental data, $O(10^5)$ instantaneous measurements per height, are clustered in 40 bins in $Z$-direction. The procedure is similar to the one described in section 4.3.1 but now only bins containing only 10 data points or less are discarded.

It was already seen in figure 4.4 that the FGM-PL and the FGM-PC database significantly overestimate H$_2$ mass fractions; from figure 4.8 it is clearly visible that this overestimation occurs under fuel-rich conditions. This can be explained by the fact that species can only diffuse in $Y$-direction in premixed flamelets. In non-premixed flamelets these species diffuse towards the $Z = Z_{stoich}$ plane where they are further oxidized [Barlow2005]. The counterflow diffusion flamelet-based databases both yield substantially more accurate predictions for H$_2$ mass fractions. Apparently the underestimation of H$_2$ mass fractions by
4.3 A priori validation of FGM databases

**Figure 4.8:** Averaged conditional \( \text{H}_2 \) mass fractions in Sandia Flame C and F at \( x=15D \) and 30D. Black circles denote measurements, red lines with square markers denote predictions from the FGM-PL, black lines with diamond markers denote predictions from the FGM-PC, blue lines with upwards directing triangle markers denote predictions from the FGM-DL and magenta lines with downwards directing triangle markers denote predictions from the FGM-DC. The blue dashed line represents the number of measurements per \( Z \)-bin; values are stated on the right \( y \)-axis. The vertical black dashed line indicates the stoichiometric mixture fraction.

**Figure 4.9:** Averaged conditional CO mass fractions in Sandia Flame C, D, E and F at \( x=15D \). Symbols identical to those in figure 4.8 have been used.

As shown in figure 4.9, the CO mass fraction tends to be overestimated by premixed

the FGM-DC at large \( \text{H}_2 \) mass fraction values, as can be seen in figure 4.4, does not have a visible effect on spatial predictions.
flamelet-based databases, although not as serious as $\text{H}_2$ mass fractions. The FGM-PL yields a larger overestimation of CO mass fractions than the FGM-PC, especially in fuel-rich regions ($Z > 0.5$). The FGM-DC can be seen to overestimate CO mass fractions in fuel-lean areas. Similar profiles are found at other heights. At each height the observation can be made that the maximum CO mass fractions decreases with increasing Reynolds number.

This phenomena is also observed for counterflow diffusion flamelets with increasing strain rate; on the hand it should be verified whether the steady-state assumption, implied by the use of a tabulation method, holds for CO. The overprediction of CO mass fractions by the FGM-PL in fuel-rich regions ($Z > Z_{st}$) and by the FGM-DC in fuel-lean regions ($Z < Z_{st}$) corresponds to an underprediction of CO$_2$ mass fractions in the same regions.

For OH it is observed that FGM-PL, the FGM-PC and the FGM-DL database practically predict the same OH mass fractions. However, the maximum OH mass fractions are overestimated by 25% at maximum. The comparison between the FGM-DC and FGM-DL shows a remarkable phenomena: near the inlet plane ($x/D=7.5$ and $x/D=15$) the FGM-DC predicts a lower maximum OH mass fraction than the FGM-DL but this maximum is located at approximately the same $Z$ as where the maximum OH mass fraction is predicted by the FGM-DL. In the far-field ($x/D=45$ and $x/D=60$) the maximum OH mass fractions are similar for both FGM databases but the maximum OH mass fraction predicted by the FGM-DC is shifted to more fuel-lean zones. Figure 4.10 shows these trends for Sandia Flame D; Flame C, E and F show a similar trend. No exact reason can be given for these phenomena but it is believed by the authors that differential diffusion of other species causes an alteration in their concentrations and thereby an alteration in the production and consumption rate of OH.

Since OH mass fractions tend to increase with increasing temperature it was put forward that the overestimation of OH might be linked to an overestimation of temperature by FGM databases. An analysis for the FGM-DL database shows that temperature is slightly underestimated, within the measurement uncertainty equal to 3% [Barlow2001], for heights up to $x/D = 45$ for all flames. When temperature is added as a control variable, i.e.
mapping is now described by $\phi = \phi(Z, Y, T)$, OH predictions do not improve and sometimes even worsen. The addition of temperature as a third control variable is therefore abandoned.

### 4.3.3 Differential diffusion effects

From previous results for $\text{H}_2$ and CO can be concluded that diffusion in $Z$-direction is of major importance for the Sandia Flames considered here. The accuracy of reproduction of differential diffusion phenomena will therefore be assessed for counterflow diffusion flamelet-based databases only. To compare the FGM-DL to the FGM-DC the differential diffusion parameter $z$ as defined by Barlow et al. [Barlow2005] is used to quantify the relative importance of molecular diffusion compared to turbulent transport. The definition of the differential diffusion parameter reads:

$$z = \frac{[Z_H - Z_{H,2}]}{[Z_{H,1} - Z_{H,2}]} - \frac{[Z_C - Z_{C,2}]}{[Z_{C,1} - Z_{C,2}]}$$

(4.19)

in which the same denotations have been used as in equation (4.2).

In figure 4.11 the differential diffusion parameter predicted by the FGM-DL and FGM-DC databases is compared to the experimentally observed differential diffusion parameter for Flame C at multiple heights. In Flame C, having only a moderate turbulence intensity, close to the inlet plane ($x=7.5D$ and $x=15D$) the FGM-DC database yields more accurate predictions for the differential diffusion parameter than the FGM-DL database. With increasing axial coordinate the FGM-DL database yields better results than the FGM-DC database.

In figure 4.12 the differential diffusion parameter predicted by the FGM-DL and FGM-DC databases is compared to the experimentally observed differential diffusion parameter.
close to the inlet plane \((x/D=7.5)\). For all flames the decrease of \(z\) for \(Z \in [0.35, 0.8]\), caused by the preferential diffusion of \(H_2\) away from this fuel-rich side of the stoichiometric condition [Barlow2005], is severely overestimated by the FGM-DC; here the FGM-DL corresponds significantly better to measurements. For Sandia Flame C it can be observed that the FGM-DC in general yields the best results although for the region \(Z \in [0.6, 1]\) the FGM-DL corresponds slightly better to measurements. This can be explained by the fact that this fuel-rich zone corresponds to an area close to the fuel jet where turbulent mixing is important. To illustrate this the turbulent kinetic energy \(k\), which is reconstructed from data by Schneider et al. [Schneider2003] is shown in the same figure. Reconstruction of \(k\) makes use of the assumption that fluctuations in tangential direction are comparable to fluctuations in radial direction: \(k = \frac{1}{2} (\overline{u'v'} + 2\overline{\nu'^2})\). Turbulent fluctuations are only available at all heights for Flame D and F. From figure 4.12 and 4.13 a significant relation can be observed between the increase of turbulent kinetic energy and a shift from the FGM-DC database to the FGM-DL database. These trends have also been observed by Barlow [Barlow2005]; this study confirms these observations by comparison with a FGM-DL and a FGM-DC database. Additionally it is showed that the departure from the
FGM-DC database towards the FGM-DL database for the region $Z \in [0.6, 1]$ is most likely caused by turbulent mixing between the fuel jet and the co-flow.

4.4 Conclusions

In this study the accuracy of FGM databases in predicting $\text{H}_2\text{O}$, $\text{H}_2$, $\text{CO}_2$, CO and OH mass fractions in partially-premixed jet flames has been assessed. Four FGM databases have been compared: databases using premixed or counterflow diffusion flamelets either with multi-component diffusion or unit Lewis numbers for all species. Both counterflow diffusion flamelet-based databases proved to be significantly more accurate than premixed-based ones for $\text{H}_2$, $\text{CO}_2$ and CO mass fraction predictions. For fuel-rich conditions the FGM-PL and FGM-PC database tend to severely overestimate $\text{H}_2$ and CO mass fractions while underestimating $\text{CO}_2$ mass fractions. Differential diffusion effects are most visible in Flame C and D and only close to the inlet plane. An increase in turbulent kinetic energy generated in the shear layer between the fuel jet and the co-flow causes the transition from a multi-component diffusion behavior to unit Lewis number diffusion. From results presented in section 4.3.1 can be concluded that an FGM-DL database, parameterized on $Z$ and $\mathcal{Y}$, can accurately predict $\text{H}_2\text{O}$, $\text{H}_2$, $\text{CO}_2$ and CO mass fractions for the Sandia Flames C to F. For OH the FGM-DL database yields a good correspondence between averages of large ensembles of measured and predicted values but for individual measurements the standard deviation is large due to measurement uncertainties (10%) and reconstruction uncertainties for $Z$ and $\mathcal{Y}$ in combination with a very localized OH peak in $Z$-$\mathcal{Y}$ space as can be seen in figure 4.1. When averaged OH mass fractions are conditioned on $Z$, the FGM-DL predicts the location of maximum OH mass fraction in $Z$-space accurately. However, the OH mass fraction at its maximum is overpredicted by approximately 25%.

Reported errors in species mass fraction predictions from RANS and LES simulations [Pitsch2000, Vervisch2004] using flamelet-based combustion chemistry tables therefore must stem rather from the subfilter model and modeling assumptions in the transport equations than from combustion chemistry representation by an FGM database provided that the choice of flamelet type and molecular transport model agree with the modeled flame. These conclusions are not surprising or contradictory to previous results but the accuracy that can be reached with a two-dimensional ($Z,\mathcal{Y}$) FGM-DL database is striking. Even for Sandia Flame F, which is known to exhibit significant extinction and re-ignition events, the predictions are in close agreement with experimental observations.
LES of Sandia Flames

In chapter 4 it has been shown that by using an FGM table, one can obtain accurate species mass fraction predictions, provided that the flamelet type and the used molecular transport model are representative for those observed in the experimental flame. In this chapter, the FGM table yielding the best results in chapter 4 is used in Large Eddy Simulations of the Sandia Flames, in order to assess the influence of errors in control variables $Z$ and $Y$ on species mass fraction predictions. These errors, originating from numerical and modeling errors in the transport equations for the control variables, were excluded in the a priori analysis in chapter 4. This chapter is focused on CO and H$_2$ mass fraction, since these were predicted less satisfactory in other studies than CH$_4$, O$_2$ and H$_2$O mass fractions.

5.1 Introduction

Direct Numerical Simulation (DNS) is impossible for nearly all industry-relevant non-premixed turbulent flames. The large variation in flame- and turbulence length scales requires a very large number of grid nodes. DNS of non-premixed flames is therefore limited to simplified flames only [Vervisch1998]. Large Eddy Simulation (LES) based methods relieve numerical requirements by filtering the DNS equations over a coarser numerical grid. This gives rise to unknown subfilter contributions for both flow and combustion chemistry which need to be modeled simultaneously. The application of LES to flame simulations receives an increasing amount of attention (see reviews [Bilger2005, Pitsch2006]). At the same time, detailed chemistry is required for accurate predictions of species mass fractions, especially for pollutant species like CO and NO. However, due to the same computational restrictions, it is unrealistic to solve transport equations for many species. To reduce computational requirements to an acceptable level while keeping all detailed chemical kinetics, the Flamelet Generated Manifold (FGM) method [Oijen2000], is one of the methods that can be used to reduce combustion chemistry.

The FGM reduction method generates a manifold, in composition space, based on one-dimensional flame structures and can therefore be considered to be a combination of classic flamelet- and manifold methods. A database is created for specific well-defined conditions without the removal of elementary reactions or species from the used chemical reaction mechanism. In this database, thermochemical variables are stored as a function of a small number of rate-controlling variables for which a transport equation is solved. This results in a large reduction in the number of required transport equations and a reduction
of the stiffness of the system of equations.

The experimentally well-documented Sandia Flame D and F [Barlow1998] will serve as test cases. The Sandia Flames have received a huge amount of attention among modelers, most probably because the extensive and conscientious collected measurement data provides the support these modelers’ need. These flames are partially-premixed methane-air ($X_{CH_4, fu} = 0.25$) jet flames supported by a premixed pilot flame, ranging from moderately turbulent (Flame D, $Re_{jet} = 22.400$) to heavily turbulent with local extinction and re-ignition (Flame F, $Re_{jet} = 44.800$). Extensive measurements of the flow field [Schneider2003] and species mass fractions [Barlow2003] enable thorough validation of numerical results. For CO, Two-Photon Laser-Induced Fluorescence [Barlow1998, Meier2000] measurements by Dally et al. [Dally2003] are used instead of Raman Scattering measurements [Dally1998]: they provide better accuracy because they suffer less from laser-excited interference from larger hydrocarbons formed on the rich side of the reaction layer.

In both Reynolds-Averaged Navier-Stokes (RANS) simulations and Large Eddy Simulations (LES) of Sandia Flame D, it has been observed that CH$_4$, O$_2$ and H$_2$O mass fractions were well predicted by premixed flamelet-based tabulated chemistry [Bradley2002, Vervisch2004, Vreman2008] or a four-step reduced reaction mechanism [Mustata2006]. However, using either a premixed flamelet-based table or a very simple reaction mechanism, in all mentioned studies it was observed that CO and H$_2$ mass fractions were predicted less satisfactorily, especially on the rich side. The a priori analysis in chapter 4 pointed out that CO and H$_2$ mass fractions from counterflow diffusion flamelets are in better correspondence with experimental data than from premixed flamelets for fuel rich conditions. The use of only a single counterflow diffusion flamelet ($a = 100$ s$^{-1}$) in an LES resulted in an improvement of CO and CO$_2$ mass fraction predictions, compared to premixed flamelet-based tabulated chemistry [Sheikhi2005] but left significant room for further development. Vreman et al. [Vreman2008] replaced the premixed flamelet-based FGM by a counterflow diffusion flamelet-based FGM, and mass fraction predictions for CO and H$_2$ were much more accurate. The fact that, although the PDF for $Z$ was very accurately reproduced when only a single counterflow diffusion flamelet was used [Sheikhi2005] CO predictions from Vreman et al. [Vreman2008] were better, suggests that for this species reaction progress is an important controlling variable. It should be noted that CH$_4$, O$_2$ and H$_2$O mass fraction predictions did not deteriorate when counterflow diffusion flamelet-based tables were used [Bradley2002, Vreman2008]: apparently predictions for these species are not sensitive to the type of flamelet which is used to generate the chemistry table.

Until now, all discussed results concerned the moderately turbulent Sandia Flame D. Based on results of previous studies [Bradley1990, Bradley1998], Bradley et al. [Bradley2002] hypothesized that premixing plays a non-negligible role in turbulent flames where quenching and re-ignition occur: in other words, premixed flamelet-based tabulated chemistry should be more appropriate for Sandia Flame F than for Flame D. Figure 5.1 shows that quenching and re-ignition are present in the rich side ($Z > 0.5$) of Flame F, since mean conditional progress variable values lay below the most strained steady counterflow-diffusion flamelet solution in this region. For Flame D quenching is almost completely absent indicated by the left graph in figure 5.1. The hypothesis from Bradley et al. was confirmed for turbulent partially premixed lifted flames [Chen2000]
where accurate predictions of lift-off heights were obtained using a Z-G approach in which $s_L$ is obtained from a premixed flamelet library. CO and H$_2$ mass fraction predictions by Bradley et al. [Bradley2002], using a premixed flamelet library, improved slightly when results for Flame F are compared to simulation outcomes for Flame D. However, Vreman et al. [Vreman2008] showed that for Flame F, like for Flame D, best CO and H$_2$ mass fraction predictions are obtained when a counterflow diffusion flamelet-based chemistry table is used instead of a premixed flamelet-based table, which is in agreement with the a priori analysis in chapter 4. Recent results from Vreman et al. [Vreman2008] and chapter 4 apparently contradict the hypothesis of Bradley et al. [Bradley2002]. Based on these results from literature, for the simulations of the Sandia Flames in this chapter a counterflow diffusion flamelet-based FGM will be used.

In this chapter can be seen as an extension of chapter 4: in LES of the Sandia Flames, errors additional to those included in the a priori analysis are now considered as well. Due to the importance of CO and H$_2$ mass fraction predictions in engineering applications, this chapter will focus on these two species. Chapter 4 has proven that when an appropriate FGM table is used, fairly good predictions for CO and H$_2$ mass fractions can be obtained in an a priori analysis. However, in the a priori analysis Z and $\gamma$ were directly reconstructed from measurements whereas LES results for these variables will contain numerical and modeling errors. The LES results will thus contain 1) errors due to chemistry representation by the FGM table, 2) interpolation errors in data retrieval from the FGM table, 3) errors due to modeling assumptions in the LES equations and prescription of their boundary conditions and, finally, 4) numerical errors in the LES due to grid resolution and accuracy of the employed numerical schemes. It appears that the latter two errors greatly influence CO and H$_2$ mass fraction predictions, as will be shown in section 5.5.3. Due to the long timescale associated with oxidation of CO and H$_2$ in these partially-premixed flames, the question rises whether mass fractions for these species should be interpolated directly from flamelet tables, or should be obtained by solving an additional transport equation. For NO, for which the formation is known to be a very slow process, it has been shown that the use of its chemical source term in a transport equation, instead of direct interpolation from the chemistry table, gives much better predictions [Vreman2008]. In order to examine whether for CO and H$_2$ a transport equation should be used, or direct interpolation from the FGM table obtains better predictions, results from these two approaches will be compared.
Chapter structure

Section 5.2 describes the equations in the LES code, their numerical discretization methods and the applied subfilter combustion model. Subsequently section 5.3 discusses the flamelet equations and how the FGM table is generated. A $\beta$-PDF subfilter closure is applied for both FGM control variables: the implementation will be explained briefly. Sandia Flame D and F are simulated: the entire numerical set-up and boundary conditions are described in section 5.4. Section 5.5 discusses LES results and, finally, conclusions from this study are summarized and discussed in section 5.6. Throughout this chapter the focus will be on CO and H$_2$ since these species are of great interest for industrial applications due to stringent environmental legislation.

5.2 LES equations and numerical discretization

In this study a FGM table is deployed to represent reaction kinetics. Thermochemical variables $\rho$, $\dot{\omega}$, $Y$, $T$ and $c_p$ are tabulated as functions of mixture fraction $Z$ describing mixing and reaction progress variable $Y$ describing reaction progress. The feasibility of the $Z$-$Y$ approach has been demonstrated in other studies of partially-premixed flames, e.g. [Huijnen2007, Pierce2004, Pitsch2000, Vreman2008]. $Z$ is defined by:

$$Z = \frac{2 M_H^{-1} [Z_H - Z_{H,2}] + 0.5 M_C^{-1} [Z_C - Z_{C,2}]}{2 M_H^{-1} [Z_{H,1} - Z_{H,2}] + 0.5 M_C^{-1} [Z_{C,1} - Z_{C,2}]}$$

(5.1)

in which $Z$ denotes element mass fractions of respectively hydrogen $H$ and carbon $C$. The subscripts fu and ox denote the fuel and oxidizer stream respectively. Definition 5.1 by Barlow et al. [Barlow2003] is a modification of the original definition proposed by Bilger [Bilger1990], equation (2.48). By removing the oxygen terms, the mixture fraction value extracted from experiments is less sensible to experimental noise and interference from laser-induced fluorescence. For the reaction progress variable $Y$ a linear combination of species mass fractions is used:

$$Y = \sum_{i=1}^{N_s} \alpha_i Y_i,$$

(5.2)

in which $\alpha_i = 0$ for all species except for CO$_2$, H$_2$O and H$_2$. Weight factors for these species are $\alpha_{CO_2} = M_{CO_2}^{-1}$, $\alpha_{H_2O} = M_{H_2O}^{-1}$ and $\alpha_{H_2} = M_{H_2}^{-1}$ which allows unambiguous mapping of the dependent variables for the Sandia Flames fuel and oxidized composition. For the definition of $Y$ given by equation (5.2), $Y_0 = 0$ for any given value of $Z$; its equilibrium value $Y_{eq}$ depends on $Z$.

The LES formulation starts from the Navier-Stokes equations with tabulated chemistry, which are identical to equations (3.1-3.4) which were introduced in chapter 3. These equations are filtered over the length scale $\Delta_r$, the filter width, which is taken equal to the local mesh-size. Filtered quantities are denoted by an overline, e.g. $\overline{\varphi}$, while Favre-Averaging (FA) is applied to velocity components, $Z$ and $Y$: i.e. $\overline{\varphi} = \overline{\varphi}/\overline{\rho}$ where $\overline{\varphi}$ can denote any velocity component, $Z$ or $Y$. The eddy-viscosity model by Vreman [Vreman2004] is used to close the nonlinear terms in momentum and scalar equations. The eddy-diffusivity for both $Z$ and $Y$ is linked to the eddy-viscosity $\mu_T$ by a fixed turbulent Schmidt number, $Sc_T = 0.6$, which is higher than the value 0.4 reported in.
5.2 LES equations and numerical discretization

studies [Goldin2005, Ihme2008-2, Pitsch2000, Vreman2008], but smaller than values 0.7 [Huijnen2007] or 1.0 [Bradley2002]. For the present grid resolution, which is significantly higher than previous studies [Pitsch2000, Vreman2008], the subfilter contribution will appear to be quite limited, thereby rendering the influence of the chosen value for $Sc_T$ very small. For both the control variables $Z$ and $Y$ unit Lewis numbers are assumed under the assumption that on the smallest resolved scales turbulent redistribution of species and enthalpy dominates molecular diffusion. Subfilter terms originating from nonlinearities in $\mu_L$, $\lambda$ and $c_p$ are neglected, which is common practice in LES [Vreman2008]. The approximated filtered equations describing turbulent combustion with tabulated chemistry become:

\begin{align}
\frac{\partial \rho}{\partial t} + \frac{\partial \rho \bar{u}_i}{\partial x_j} &= 0, \\
\frac{\partial \rho \bar{u}_i}{\partial t} + \frac{\partial \rho \bar{u}_i \bar{u}_j}{\partial x_j} &= \frac{\partial \rho}{\partial x_i} \left[ (\mu_L + \mu_T) \left( \frac{\partial \bar{u}_i}{\partial x_j} + \frac{\partial \bar{u}_j}{\partial x_i} - \frac{2}{3} \frac{\partial \bar{u}_k}{\partial x_k} \delta_{ij} \right) \right], \\
\frac{\partial \rho \bar{Z}}{\partial t} + \frac{\partial \rho \bar{u}_i \bar{Z}}{\partial x_j} &= \frac{\partial \rho}{\partial x_i} \left[ \frac{\lambda}{c_p} \frac{\mu_T}{Sc_T} \frac{\partial \bar{Z}}{\partial x_j} \right], \\
\frac{\partial \rho \bar{Y}}{\partial t} + \frac{\partial \rho \bar{u}_i \bar{Y}}{\partial x_j} &= \frac{\partial \rho}{\partial x_i} \left[ \frac{\lambda}{c_p} \frac{\mu_T}{Sc_T} \frac{\partial \bar{Y}}{\partial x_j} \right] + \bar{\omega}_Y,
\end{align}

where the summation convention is used over indices $j$ and $k$ and a Newtonian behavior of the fluid is assumed. $\bar{p}$, $\bar{\omega}_Y$, $\bar{T}$ and $c_p$ are retrieved from the FGM table using linear interpolation in both $Z$ and $Y$ direction. The effective dynamic viscosity $\mu$ is composed of a turbulent and molecular contribution: $\mu = \mu_T + \mu_L$. Simplified formulations for $\mu$ and $\lambda$ for CH4-air flames are used [Smooke1991] to reduce computational cost:

\begin{align}
\mu_L/c_p &= 1.67 \times 10^{-8} \left( T/298 \right)^{0.51}, \\
\lambda/c_p &= 2.58 \times 10^{-5} \left( T/298 \right)^{0.69}.
\end{align}

A standard finite volume method is employed on a staggered Cartesian mesh. For the momentum equations second-order central differencing and an explicit hybrid timestepping method are applied: convective terms are integrated in time using a third-order Adams-Bashforth scheme while for viscous terms and source terms a forward Euler scheme is used. The hybrid timestepping method is formally only first-order accurate but provides significantly better stability than a pure Adams-Bashforth or a pure forward Euler method. The contribution, and thereby also the error contribution, of the viscous terms scales with the inverse of the Reynolds number when compared to the convective terms. Therefore, for turbulent flames as considered in this study in which $Re = O(10^2)$, the viscous terms can be discretized using a lower order method than the convective terms without sacrificing too much numerical accuracy. For the scalars $Z$ and $Y$ the Van Leers third-order accurate MUSCL scheme, which is TVD, is applied to the advective terms while second-order central differencing is applied to the viscous terms. The variable density approach, which involves a Poisson equation for the pressure, has been successfully applied previously by Pitsch and Steiner [Pitsch2000] and Pierce and Moin [Pierce2004]. Because the Poisson-solver is sensitive to density fluctuations, the density is filtered explicitly over twice the local mesh-size for additional numerical stability [Vreman2008]. The multigrid implementation in the code used for this study is
thoroughly described in [Vreman2009-1]. The code has been parallelized using the MPI protocol, effectively dividing the computational domain in 32 subdomains, and the OpenMP protocol.

### 5.3 Combustion modeling

Chemical kinetics are modeled by flamelet-based tables. As pointed out in chapter 4, an FGM based on counterflow diffusion flamelets provides best results for mass fraction predictions, and will therefore be used in this chapter. It will be assessed whether subfilter modeling for combustion has a significant influence: for thermochemical quantities retrieved from the FGM table, a laminar database and a database with a \( \beta \)-PDF subfilter closure for control variables \( Z \) and \( Y \) will be compared.

The equations describing quasi-steady counterflow diffusion flamelets with unit Lewis numbers for all species have been derived in section 2.2.2 but will be restated here with unit Lewis numbers to be perfectly clear. They read:

\[
\frac{\partial \rho}{\partial t} + \frac{\partial \rho u}{\partial x} = -\rho G ,
\]

\[
\frac{\partial \rho Y_i}{\partial t} + \frac{\partial \rho u Y_i}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda \partial Y_i}{c_p \partial x} \right) + \dot{\omega}_i - \rho Y_i G , \quad i \in [1, N_s - 1] ,
\]

\[
\frac{\partial \rho h}{\partial t} + \frac{\partial \rho u h}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda \partial h}{c_p \partial x} \right) - \rho h G ,
\]

\[
\rho \frac{\partial G}{\partial t} + \rho u \frac{\partial G}{\partial s} = \frac{\partial}{\partial s} \left( \mu \frac{\partial G}{\partial s} \right) + P(t) - \rho G^2 ,
\]

\[
\rho = p_0 M / (RT) ,
\]

in which the Low Mach-number approximation is used in the thermal equation of state (5.13) with \( p_0 \) denoting the ambient pressure. \( G \) denotes the strain rate [DixonLewis1990, Stahl1991], defined as

\[
G(x, t) \equiv \frac{\partial v}{\partial y} ,
\]

which is introduced to account for the \( y \)-component of the flow in a counterflow geometry. The derivation of the transport equation for \( G \), equation (5.12), is given in appendix A. In counterflow diffusion flamelets, Dirichlet boundary conditions for \( Y_i \) and \( h \) are imposed in both the oxidizer \( (x \rightarrow -\infty) \) and fuel \( (x \rightarrow \infty) \) stream while for \( G \) a combination of Dirichlet and Neumann boundary conditions is imposed:

\[
Y_i (x \rightarrow -\infty) = Y_{i,ox} , \quad h (x \rightarrow -\infty) = h_{ox} , \quad G (x \rightarrow -\infty) = a (t) ,
\]

\[
Y_i (x \rightarrow \infty) = Y_{i,fu} , \quad h (x \rightarrow \infty) = h_{fu} , \quad \frac{\partial G}{\partial x} (x \rightarrow \infty) = 0 .
\]

The translational degree of freedom is removed by choosing the stagnation plane at \( x = 0 \). The parameter of this system is the applied strain rate \( a(t) \), which is defined at the oxidizer side \( (x \rightarrow -\infty) \). Tabulated polynomial functions for \( h \) and \( c_p \) [Kee1986] and the same simplified formulation for \( \lambda \), equation (5.8), as used in LES equations (5.5-5.6) is used to close the flamelet equations. Unit Lewis numbers are used for all species for consistency with the DNS equations (5.5-5.6). Source terms for species are computed using the GRI 3.0
reaction scheme [GRI mech 30]: this reaction mechanism showed best agreement with experimental data when compared to six other (reduced) reaction mechanisms [Cao2005]. Only NO was better predicted by the GRI 2.11 reaction scheme [GRI mech 211], which is typically overestimated by a factor of two by the GRI 3.0 reaction scheme. In this study, the interest is focussed on CO and H$_2$ and therefore the GRI 3.0 reaction scheme is used. The mass fraction of the abundant species N$_2$ is determined using the identity $\sum Y_i = 1$.

Due to the employed definition for $Y$, given by equation (5.2), progress variable equilibrium values $Y_{eq}$ depend on $Z$ since the equilibrium species mass fractions on which $Y$ is based depend on $Z$. Flamelet solutions are tabulated as a function of the control variables $Z$ and $C$ of which the latter is a normalized value of $Y$ according to:

$$C = \frac{Y - Y_0 (Z)}{Y_{eq} (Z) - Y_0 (Z)} \in [0, 1].$$

(5.17)

$Y_i$ and $T$ from the flamelets are interpolated on an equidistant $Z$-$C$ grid and $\rho$ and $c_p$ are recalculated to keep them consistent with interpolated $Y_i$ and $T$ values. The equidistantly-spaced square $Z$-$C$ grid enables the use of fast interpolation routines since entry indices and weight factors can be readily computed. Approximately 400 flamelets are computed with varying strain rate $a$ starting from a steady solution with $a = 2$ [1/s]. The initial fuel temperature equals $T_{0, fu} = 294$ K and the initial oxidizer temperature equals $T_{0, ox} = 291$ K. The ambient pressure equals $p_0 = 100.6$ kPa (= 0.993 atm). From the flamelets an FGM with an equidistant ($201 \times 201$) $Z$-$C$ grid is generated for simulations without a subfilter combustion model. During an LES of a flame, $\dot{Y}$ from equation (5.6) is scaled to $C$ whenever $\rho$, $\dot{\omega}$, $T$ and $c_p$ are retrieved from the FGM database and passed to the flow solver. As an example $\dot{\omega}$ and $T$ from the FGM table are depicted in figure 5.2.

In non-premixed flames reactants have to mix prior to combustion, thereby limiting the overall consumption of reactants by the species molecular diffusion towards the flame front. In LES where the subfilter scales are not resolved, subfilter turbulence and molecular diffusion together determine the states in composition space that can be accessed through mixing. Probability Density Function (PDF) methods focus on the statistical properties of intermediate states rather than deterministic properties, which allows a straightforward description of thermochemical variables. A Favre-Averaged (FA) quantity is then obtained by:

$$\bar{\varphi} = \int \int \varphi (Z, C) \tilde{P} (Z, C) \, dZ \, dC,$$

(5.18)
in which $\tilde{P}(Z, C)$ denotes the joint-PDF which statistically describes the non-resolved local state in composition space. In flamelet regimes, the flame structure is at least partly hidden in subfilter scales and since the PDF-approach is a statistical one, it does not guarantee that the non-resolved reactive layer resembles the one observed in flamelets. Nevertheless, when the Karlovitz number indicates that turbulent eddies can not significantly influence the reaction layer, it is assumed that flamelet-like flame structures are hidden in subfilter scales. A possible approach to obtain the local joint PDF is to derive transport equations for the probability of all possible realizations of the joint-PDF [Pope1985, Haworth2010]. The large set of differential equations for the probability of occurrence of a specific combination of values for thermochemical variables, results in very high computational costs. In this study a PDF-approach is used, but instead of solving transport equations for the probability of all possible realizations the joint-PDF is modeled by a presumed shape, resulting in the Presumed PDF (PPDF) approach. In this method the joint-PDF, which is a function of only $Z$ and $C$ in this tabulated chemistry approach, is described by the first few moments of these control variables. These moments are obtained by either solving transport equations or by algebraic closures.

Simulations are performed in- and excluding a subfilter combustion term modeled by a presumed $\beta$-PDF approach. The main advantage of the $\beta$-PDF over other PDF shapes, is that it is bounded between zero and unity like the control variables $Z$ and $C$. It is expected that for Flame F, in a simulation with an identical $\Delta F$, the subfilter contribution is more important than for Flame D. It is assumed that $Z$ and $C$ are statistically independent, thereby the joint PDF in equation (5.18) can be reduced to the product of two marginal PDF’s:

$$\tilde{P}(Z, C) = \tilde{P}(Z) \tilde{P}(C) \quad .$$

In the presumed $\beta$-PDF approach, both marginal PDF’s are assumed to be fully described by their first two moments, i.e. their mean and variance:

$$\tilde{P}(Z) = \tilde{P} \left( Z; \bar{Z}, \bar{Z}^2 \right), \quad \text{and} \quad \tilde{P}(C) = \tilde{P} \left( C; \bar{C}, \bar{C}^2 \right) \quad .$$

This implies that the two-dimensional FGM table is extended with two dimensions: the FA mixture fraction variance $\bar{Z}^2$ and the FA progress variable variance $\bar{C}^2$. These quantities have to be computed by the CFD solver. This approach has been successfully applied in LES by several others, e.g. [Huijnen2007, Kempf2005, Ihme2005, Pierce2001].

Thermochemical parameters in the FGM table are now mapped on four control variables: 

$$\varphi = \varphi \left( \bar{Z}, \bar{Z}^2, \bar{C}, \bar{C}^2 \right) \quad \text{in which } \varphi \text{ can denote any Reynolds-Averaged (RA) or Favre-Averaged (FA) thermochemical variable. Manifold entries for non-zero variance values are computed using a $\beta$-distribution [Abramowitz1965] for both $Z$ and $C$. RA and FA variables are obtained by integration of properties from flamelets with the PDF’s of the control variables:}$$

$$\varphi = \bar{p} \int \int \left( \frac{\varphi(Z, C)}{\rho(Z, C)} \right) \tilde{P} \left( Z; \bar{Z}, \bar{Z}^2 \right) \tilde{P} \left( C; \bar{C}, \bar{C}^2 \right) \, dZ \, dC \quad ,$$

$$\tilde{\varphi} = \int \int \varphi \left( Z, C \right) \tilde{P} \left( Z; \bar{Z}, \bar{Z}^2 \right) \tilde{P} \left( C; \bar{C}, \bar{C}^2 \right) \, dZ \, dC \quad ,$$

in which the RA density is defined by:

$$\bar{p} = \left[ \frac{1}{\bar{\rho}} \right]^{-1} = \left[ \int \int \tilde{P} \left( Z; \bar{Z}, \bar{Z}^2 \right) \tilde{P} \left( C; \bar{C}, \bar{C}^2 \right) \, dZ \, dC \right]^{-1} \quad .$$
5.4 Numerical setup for Sandia Flame D and F

Variance entries are exponentially clustered near zero for sufficient resolution in this region. An algebraic gradient model, similar to the gradient model for the turbulent stresses, is used to estimate \( \overline{\varphi'^2} \) where \( \varphi \) denotes \( Z \) or \( Y \):

\[
\overline{\varphi'^2} = \alpha \Delta x^2 \left( \frac{\partial \varphi}{\partial x_j} \right)^2,
\]

where \( \alpha \) is assumed to be constant and the summation convention is used over index \( j \). For an equidistant grid spacing and smooth scalar fields a Taylor-expansion yields \( \alpha = 1/12 \) but previous DNS results for turbulent subfilter stress models [Clark1979] or subfilter scalar variances [Branley2001] pointed out that this value underestimates variances. In this study \( \alpha = 0.125 \) is used, which complies with values used in other studies ranging from \( \alpha = 0.1 \) [Branley2001] to \( \alpha = 0.3 \) [Vreman2008].

Due to computer memory restrictions, PDF-integrated tables use a resolution of \((101 \times 11 \times 101 \times 11)\) grid is used. Variances are quadratically clustered near zero where thermochemical variables are most sensitive to variation in variances as is depicted in figure 5.3.

For species with slow formation/consumption processes, like CO, it is not assured that they would remain on the chemical manifold if detailed chemistry would be used. Solving a transport equation for these species, instead of direct interpolation from the FGM table, introduces additional freedom for these species: they are no longer fully determined by \( Z \) and \( Y \) but by the history of these controlling variables as well. For NO, Vreman et al. [Vreman2008] have shown that the use of its source term \( \dot{\omega}_{NO} \) in an additional transport equation for NO, yields much better mass fraction predictions. With a unity Lewis number, the approximated filtered transport equation for a species \( i \) reads:

\[
\frac{\partial \rho \dot{Y}_i}{\partial t} + \rho \frac{\partial \dot{u}_j Y_i}{\partial x_j} = \frac{\partial}{\partial x_j} \left[ \left( \frac{\lambda}{S c_T} \right) \frac{\partial \dot{Y}_i}{\partial x_j} \right] + \dot{\omega}_i,
\]

in which \( Y_i \) can denote any species mass fraction for which this differential equation is solved. It will be verified for CO and H\(_2\) whether solving a transport equation yields better results than direct species mass fraction interpolation from the FGM table. It should be noted that using this approach, a species source term is a function of \( Z \) and \( Y \) only, and is therefore insensitive to its own mass fraction value directly. Non-physical mass fraction values might thus be obtained since the mass fraction value is allowed to leave the manifold and receives no feedback from its mass fraction value.

5.4 Numerical setup for Sandia Flame D and F

Sandia Flame D and F are piloted co-axial CH\(_4\)-air flames in which the fuel jet (diameter \( D = 7.2 \text{ mm}, X_{CH_4} = 0.25, X_{air} = 0.75, T_0 = 294 \text{ K} \)) is surrounded by a pilot jet in chemical equilibrium (diameter 2.62 \( D, \phi = 0.77, T = T_{eq} \)). The pilot jet is girded by a co-flow of air \( T_0 = 291 \text{ K} \). Characteristic flow velocities including the jet Reynolds number \( Re_{jet} \), for both Flame D and F are summarized in table 5.1. Velocity profiles [Schneider2003] and species mass fractions [Barlow2003, Dally2003] have been accurately measured for both flames: mass fractions of nine species, O\(_2\), CH\(_4\), N\(_2\), H\(_2\)O, CO\(_2\), H\(_2\), CO, OH and NO, are available. The sum of all measured mass fractions is less than 0.2% away from unity for all instantaneous measurements at all probed locations, implying that mass fractions of species which have not been measured do not have a significant influence on measured
quantities. The two species on which this chapter focuses, H$_2$ and CO, both have a measurement uncertainty equal to 10%.

The computational domain starts one nozzle diameter ($D$) downstream of the nozzle, where velocity and species measurements serve as inlet boundary condition. A rectangular numerical grid with an approximate size of $32D \times 32D \times 86D$ of which the last is the streamwise direction. The cells with center $x = y = 0$ were on the fuel jet axis, referred to as centerline from here on. The resolution of the numerical simulations is chosen as fine as computational resources allow, in order to minimize both numerical and modeling errors. The grid was stretched in all three directions and contained $164 \times 164 \times 484$ cells, with a minimum grid spacing of $D/19.2 = 375 \, \mu m$ near the fuel jet. This minimum grid spacing corresponds to half the estimated probe volume [Barlow1998] used to obtain experimental data. For the estimated probe volume of $750 \, \mu m$ in experiments, Barlow et al. [Barlow1998] state that spatial averaging only has a limited influence: it is therefore expected that subfilter combustion modeling will have only a slight influence on numerical results. Since the used numerical grid is Cartesian, cylindrical (radial) coordinates of cell centers are computed at the inflow plane to allow the use of flow and species mass fractions as boundary conditions. A correction factor is computed and applied to obtain exactly the bulk flow velocities for the fuel and pilot jet as they were experimentally obtained; this correction factor was smaller than 2.5% for both Flame D and F. Turbulent inflow conditions are generated using a random noise generator: for each velocity component, every timestep random numbers between $-1$ and $1$ are generated on the mesh one grid cell level below the inflow plane. A box-filter of $\Delta F = D$ in all three spatial directions and a corresponding temporal filter are subsequently applied; the temporal filter is discretized by

$$
\hat{f}_{n+1} = \left(1 - \frac{\bar{w}_0 \Delta t}{\Delta F}\right) \hat{f}_n + \left(\frac{\bar{w}_0 \Delta t}{\Delta F}\right) f_n,
$$

in which $f_n$ denotes the random number vector which is updated every timestep for each velocity component. $\hat{f}_n$ denotes the filtered velocity perturbation vector at instant $n$ and $f_{n+1}$ denotes the new filtered velocity perturbation vector at instant $n + 1$. $\bar{w}_0$ denotes the mean streamwise velocity at $x = y = 0$. The perturbation vector is initialized with $\hat{f}_0 = 0$ and multiplied by an appropriate amplification factor to ensure that $\hat{f}$ has the same amplitude as the random number generator. Finally, since $\hat{f}$ has been a normalized quantity up to now, it is multiplied by measured RMS values to obtain the required inflow
turbulence intensities. Inflow boundary conditions for \((Z,Y)\) are prescribed from experimental data; \(Y\) is computed from this data set according to equation (5.2). At boundaries in both \(x\) and \(y\)-direction and at the outflow-plane in \(z\)-direction, Neumann boundary conditions are applied for velocity components and transported scalars. A Dirichlet \((p = p_0)\) boundary condition is applied in \(x\) and \(y\)-direction while a Neumann boundary condition for the pressure is applied at the inflow and outflow plane. A constant time step of \(\Delta t = 0.6 \, \mu s\) is used for all simulations in order to keep the CFL-number sufficiently low.

**Statistical analysis**

Time-averaging of statistics was started at \(t = 0.049\) s for Flame D and at \(t = 0.022\) s for Flame F (\(\approx 1.5\) throughflow times) and simulations were continued until \(t = 0.15\) s and \(t = 0.075\) s respectively (\(\approx 4.5\) throughflow times). Compared to the throughflow time the total simulation time does not seem extremely long, but it should be noted that the computational domain is approximately two flame heights in streamwise direction. Comparison of the final statistics with statistics obtained approximately one throughflow time earlier, proved the statistics to be converged. Autocorrelation functions for the streamwise velocity component, the mixture fraction and the progress variable at the centerline are presented in figure 5.5. From these autocorrelations, correlation times are obtained by fitting the exponential function \(R = \exp \left(-t/\tau\right)\). In Flame D, mixture fraction and progress variable had slightly larger correlation times then the streamwise velocity, \(\tau \approx 0.15\) ms at \(z/D = 30\) and \(\tau \approx 0.32\) ms at \(z/D = 45\). This definition for correlation times results in slightly smaller values than obtained by Huijnen [Huijnen2007], but a comparable trend is obtained when results at different heights are compared. The obtained
correlation time at $z/D = 45$ is approximately hundred times smaller than one
throughflow time, implying that the total statistics gathering time, $3.0$ throughflow times
$\approx 300\tau$, should be more than sufficient to obtain converged statistics. For Flame F, mixture
fraction and progress variable correlation times equal $\tau \approx 0.11$ ms at $z/D = 30$ and
$\tau \approx 0.29$ ms at $z/D = 45$, which are slightly smaller values than obtained for Flame D. The
obtained correlation time at $z/D = 45$ for Flame F is approximately fifty times smaller than
one throughflow time, implying that for this flame the statistics gathering time $\approx 150\tau$.
This should ensure converged statistics for Flame F as well as Flame D.

5.5 Results

Presented results are split in three parts: first parameters concerning numerical aspects of
the simulation are discussed and both flames are characterized in terms of length scales.
Subsequently simulation outcomes for the controlling variables $\tilde{Z}$ and $\tilde{Y}$ are discussed
since they determine all thermochemical variables which are retrieved from the FGM
tables. An instantaneous snapshot of the controlling variables and their variance in the
$xz$-plane intersected by the centerline, is presented for Sandia Flame F in figure 5.6.
Dependent variables, mass fractions for CO and H$_2$ on which is focussed in this study, are
finally presented; a instantaneous snapshot of $\tilde{Y}_{CO}$ for Sandia Flame F is shown as well in
figure 5.6. In this figure, it can be seen that CO is produced on the rich side of the
stoichiometric plane, and that it is subsequently consumed by oxidation with entrained air
from the co-flow.
5.5 Results

Figure 5.6: Snapshot of Sandia Flame F at 49.50 ms, which corresponds to approximately 3.5 throughput times of the computational domain. From top left to bottom right the FA mixture fraction $\tilde{Z}$, the FA mixture fraction variance $\tilde{Z}'^2$, the FA progress variable $\tilde{Y}$, the FA progress variable variance $\tilde{Y}'^2$, the RA density $\tilde{\rho}$ and the FA CO mass fraction $\tilde{Y}_{\text{CO}}$ are depicted; colors indicate their values. The black iso-contour indicates the stoichiometric $Z$-value ($Z_{\text{st}} = 0.351$).

5.5.1 Subfilter modeling evaluation

The contribution of the subfilter model for momentum transport is quantified by comparing the subfilter eddy viscosity $\mu_T$ with the molecular viscosity $\mu_L$. From figure 5.7 it can be seen that for both Flame D and F subfilter modeling for momentum transport is
important close to the nozzle exit, while further downstream the flow is properly resolved, since there the molecular viscosity is greater than the subfilter eddy viscosity. As expected, the absolute subfilter contribution in Flame F is larger than for Flame D due to the higher turbulence intensity. For both flames, the subfilter contribution for momentum transport can not be neglected in the near-jet region, where the shear layer between the central fuel jet and the pilot jet generates turbulence. The in- or exclusion of the subfilter combustion model does not significantly alter results. Apparently, subfilter fluctuations in density and temperature, which determines the diffusion coefficient $\lambda/c_p$, do not have a significant influence on the resolved flow field.

Figure 5.8 shows that subfilter modeling for combustion has a small but non-negligible influence on the profile for the time-averaged progress variable source term $\overline{\omega Y}$. This is consistent with previous results from Vreman et al. [Vreman2008] for Flame D, who used a more than twice as coarse numerical grid: $\Delta_x = 0.9$ mm compared to $\Delta_x = 0.375$ mm for this study, and also found only a minor influence on progress variable profiles. The subfilter closure for combustion is retained for further simulations: it is expected that its influence is non-negligible for thermochemical variables which exhibit a non-linear behavior with respect to the control variables, like mass fractions of CO and H$_2$.

The Kolmogorov length scale $\eta$, the streamwise Taylor length scale $\lambda_T$ [Vreman2008] and
## 5.5 Results

### Bulk flow velocities

<table>
<thead>
<tr>
<th></th>
<th>Flame D</th>
<th>Flame F</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel jet [m/s]</td>
<td>49.6 / 22.400</td>
<td>99.2 / 44.800</td>
</tr>
<tr>
<td>Pilot jet [m/s]</td>
<td>11.4</td>
<td>22.8</td>
</tr>
<tr>
<td>Co-flow [m/s]</td>
<td>0.9</td>
<td>0.9</td>
</tr>
</tbody>
</table>

### Kolmogorov length scale

| $\eta$ [m] $(z = 30D/45D)$ | $1.10 \times 10^{-4}$ / $2.59 \times 10^{-4}$ | $1.13 \times 10^{-4}$ / $2.01 \times 10^{-4}$ |

### Taylor length scale

| $\lambda_T$ [m] $(z = 30D/45D)$ | $1.53 \times 10^{-3}$ / $3.23 \times 10^{-3}$ | $2.05 \times 10^{-3}$ / $3.07 \times 10^{-3}$ |

### Taylor length scale Reynolds number

| $Re_{\lambda_T}$ $(z = 30D/45D)$ | 70.1 / 53.3 | 98.1 / 68.1 |

| $Ka$ $(z = 30D/45D)$ | 82.6 / 14.9 | 78.3 / 24.75 |

Table 5.1: Characteristic numbers for Sandia Flames D and F.

Taylor scale Reynolds number $Re_{\lambda_T}$ are defined by

\[
\eta = \left( \frac{\nu^3}{\epsilon} \right)^{1/4}, \tag{5.27}
\]

\[
\lambda_T = \sqrt{\frac{u'w'/(\partial u'/\partial z)^2}{\epsilon}}, \tag{5.28}
\]

\[
Re_{\lambda_T} = \frac{w'\lambda_T}{\nu^{-1}}, \tag{5.29}
\]

in which $\nu = \nu_T + \nu_L$ denotes the effective kinematic viscosity, containing a turbulent and molecular contribution. $\epsilon$ denotes the viscous dissipation and $u'$ denotes the streamwise velocity fluctuation. Definition (5.28) corresponds to the transverse Taylor length scale deduced by Pope [Pope2000]. For the streamwise Taylor length scale Pope found an additional factor $\sqrt{2}$ but this factor is omitted here since equation (5.28) yields a mere estimate for $\lambda_T$. From table 5.1 it can be seen that $\eta$ is of the same order of magnitude as the minimal filter width ($3.75 \times 10^{-4}$ m) in the region where the flame brush crosses the centerline: $30D \lesssim z \lesssim 45D$. To determine an estimate for the Karlovitz number, $Ka = (\delta_L/\eta)^2$, the flame thickness $\delta_L$ is derived from laminar counterflow flames. $\delta_L$ is based on the temperature increase from oxidizer temperature, ($= 291$ K), to the maximum temperature and the maximum temperature gradient. A counterflow flamelet with $a = 100$ [1/s] has a flame thickness of $\delta_L = 1.642$ mm while the most strained steady flamelet solution that can be obtained for the boundary conditions corresponding to the Sandia Flames, having a strain rate equal to $a = 978$ [1/s], has a flame thickness of $\delta_L = 0.497$ mm. To derive Karlovitz numbers, of which mainly the order of magnitude is important, the average flame thickness is approximated to be $\delta_L = 1.0$ mm. Seshadri and Peters [Seshadri1988] state that the flamelet concept is valid for $Ka \leq O(10^2)$ which indicates that, given the obtained Karlovitz numbers tabulated in table 5.1, even in Flame F flamelet-based chemistry tables are applicable. The comment that is to be made here is that the Karlovitz number depends on the squared flame thickness and is therefore very sensitive to the assumed value for $\delta_L$. When the value for $\delta_L$ for the most strained steady flamelet solution is used instead, the Karlovitz number limit mentioned by Seshadri and Peters [Seshadri1988] is exceeded.
5.5.2 Flow field and transported scalars

Flow field

Mean streamwise velocity $\bar{w}$ and turbulent kinetic energy $k$ results are shown in figures 5.9 and 5.10 and compared with measurements from Schneider et al. [Schneider2003]. Reconstruction of $k$ from measurements makes use of the assumption that fluctuations in tangential direction are comparable to fluctuations in radial direction: $k = \frac{1}{2} (\overline{w'd'} + 2\overline{v'v'})$. The centerline results indicate that the spreading of the jet is underestimated for both Flame D and F. This is conformed by the radial profiles which consistently show an overestimation of the streamwise velocity at the centerline and an underestimation in the far-field. A well-known modeling issue for these flames, is the thin mixing boundary layer between the fuel jet and pilot flow. Even with the currently used resolution, which is approximately twice as high as the resolution used by Pitsch and Steiner [Pitsch2000] and Vreman et al. [Vreman2008], this thin layer is hard to resolve and leads to $k$-values which rise too strong around $z/D = 15$ for Flame F. This too sharp rise in $k$ has been reported by Kempf et al. [Kempf2005] and Huijnen [Huijnen2007] as well. To arrive at reasonable $k$-values in the flame brush, the prescribed amplitude of the turbulent fluctuations in the inflow plane has been deliberately decreased below experimentally observed values. At heights $z/D = 15$, $z/D = 30$ and $z/D = 45$ turbulent kinetic energy values are fairly well recovered by the simulation. The region $z/D < 15$ in which $k$-values are severely underestimated, is the most probable cause for the underestimated spreading of the jet. This is verified in a reference computation for Flame F (dash-dotted line in right graph of figure 5.9) in which $k$ was kept constant at the inlet while the filter width for turbulent fluctuations at the inflow plane is reduced by a factor 8, effectively reducing the turbulent integral length scale and thereby reproducing a larger part of the turbulent spectrum. This reduction of the turbulent integral length results in an earlier transition of the shear layer and an associated earlier spreading of the jet. In RANS simulations of Sandia Flame D, a similar sensitivity to the prescription of turbulence at the inflow plane has been observed [Merci2002]. The current observations do not coincide with previous comments of Pitsch and Steiner [Pitsch2000] and Vreman et al. [Vreman2008] who found only a small sensitivity to inflow turbulence intensities.

Concluding, it can be stated that the mean streamwise velocity field and turbulence intensities are not very well recovered in the current simulations. This is most probably caused by the prescription of the turbulent inflow conditions which should have a higher turbulence intensity and, most probably, a smaller integral length scale to cause a more downstream turbulent break-up of the central jet. It is observed that the flow field of Flame F is affected less by the error in the prescribed turbulent perturbations than Flame D: apparently the overshoot in $k$ partly compensates its underestimation at the inflow boundary.

Transported scalars

In the current FGM chemistry approach, transport equations (5.5) and (5.6) are solved for $Z$ and $\chi$. All other required thermochemical variables like $\rho$, $\omega_Y$, $T$, $c_p$ and species mass fractions are retrieved from the FGM table. This implies that good species mass fraction predictions require a good correspondence of $Z$ and $\chi$ from simulations with experiments; if this is not the case, wrong regions of the FGM table are accessed which will not lead to an accurate reproduction of experimental results.
5.5 Results

Figure 5.9: Centerline mean streamwise velocity and centerline turbulent kinetic energy. Upper lines denote centerline streamwise velocity from simulations and open symbols denote velocity measurements. Lower lines denote centerline turbulent kinetic energy and closed symbols their experimentally obtained values. Solid lines represent simulations with subfilter combustion model, dashed lines represent simulations in which $\dot{\omega}_Y$ is approximated by $\omega_Y$. The dash-dotted line depicted in the right graph represents a reference simulation without subfilter combustion model and an inflow filter width for turbulent fluctuations of $\Delta_F = D/8$ instead of $\Delta_F = D$.

Figures 5.11 and 5.12 depict axial and radial mixture fraction profiles together with experimental results of Barlow et al. [Barlow2003]. Since $Z$ is a passive scalar, profiles of $Z$ are very similar to those of the velocity field which dictates mixing processes. For both Flame D and Flame F, the underestimated values for $k$ close to the nozzle exit result in a $Z$-field which is slightly shifted downstream. At heights $z/D = 30$ and $z/D = 45$ the mixture fraction RMS values are well recovered, but in the upstream region also comprising $z/D = 15$ it is underestimated due to the underestimated turbulent kinetic energy. In the inflow boundary conditions for $Z$, no fluctuations are included. This explains why the computed mixture fraction RMS value is negligible close to the nozzle exit, contrary to experimental data. In simulations of Pitsch and Steiner [Pitsch2000], Vreman et al. [Vreman2008] and Huijnen [Huijnen2007] similar simulation results have been obtained. The most probable cause for the experimentally observed mixture fraction RMS value close to the nozzle exit are the errors in the experimental technique [Pitsch2000], since the RMS value has to go to zero close to the nozzle exit. Like for the mean streamwise velocity, results for Flame F exhibit a better correspondence with experiments than Flame D. The inclusion of the subfilter combustion model does not significantly affect profiles of this passive scalar.

In figures 5.13 and 5.14 axial and radial profiles of the reaction progress variable $\gamma$ are depicted together with values reconstructed from species measurements [Barlow2003] and equation (5.2). The progress variable variance is reconstructed by computing the progress variable from a large number of instantaneous measurements; the required measurement data is available only for the radial profiles. Contrary to results for the flow field and mixture fraction, simulation results for Flame D and Flame F exhibit a different behavior. For Flame D the time-averaged maximum $\gamma$-value is predicted downstream of the measured maximum value while for Flame F this maximum value is predicted upstream of the experimental results. Radial profiles at $z/D = 30$ confirm this different behavior: the centerline ($r/D = 0$) time-averaged $\gamma$-value of Flame F is more than twice the value obtained in Flame D while experimental results show an opposed trend. Differences between the simulation including the subfilter combustion model and the one without are
Figure 5.10: Radial profiles of mean streamwise velocity and turbulent kinetic energy at heights $z/D = 15$, $z/D = 30$ and $z/D = 45$. Upper lines denote streamwise velocity from simulations; open symbols denote velocity measurements. Lower lines denote turbulent kinetic energy; closed symbols their experimentally obtained values. Solid lines represent simulations with subfilter combustion model, dashed lines represent simulations in which $\dot{\omega}_Y$ is approximated by $\dot{\omega}_Y$.

mainly seen in the downstream region $z/D \leq 30$. From figures 5.11 and 5.12 it was already observed that this also is the region where the inclusion of the subfilter model for chemical kinetics causes deviations in the $Z$-field.

In order to separate errors in $\tilde{Y}$-profiles arising from the erroneous representation of the mixing field and errors originating from the tabulated chemistry approach, $\tilde{Y}$-profiles are presented in mixture fraction space in figures 5.15 and 5.16. High $Z$-values correspond to small values of $z/D$ or $r/D$, which is physically located close to the nozzle exit or the centerline. Small values of $Z$ correspond to the far-field. From the axial profiles and the radial profiles at height $z/D = 15$ it can be concluded that the chemical source term for the progress variable, $\tilde{\omega}_Y$, is overestimated for high $Z$-values in both flames, although this overestimation is more severe for Flame F. A possible explanation is that the quenching and re-ignition processes which are present in Flame F, can not be adequately modeled using an FGM table based on quasi-steady counterflow diffusion flamelet solutions. This explanation is backed by the observation that the centerline $\tilde{Y}$-profile (in $Z$-space) in Flame D, which does exhibit negligible local quenching, is fairly well recovered in the LES. In simulations of unsteady counterflow diffusion flamelets in which the applied strain rate
5.5 Results

Figure 5.11: Centerline FA mixture fraction and centerline FA mixture fraction RMS values. Upper lines denote centerline FA mixture fraction from simulations and open symbols denote measurements. Lower lines denote centerline FA mixture fraction RMS values and closed symbols their experimentally obtained values. Solid lines represent simulations with subfilter combustion model, dashed lines represent simulations in which $\dot{\omega}_Y$ is approximated by $\dot{\omega}_Y$. The dash-dotted profile represents the time-average of the estimated subfilter variance by equation (5.24).

Figure 5.12: Radial profiles of FA mixture fraction and centerline FA mixture fraction RMS values at heights $z/D = 15$, $z/D = 30$ and $z/D = 45$. Upper lines denote FA mixture fraction from simulations; open symbols denote measurements. Lower lines denote FA mixture fraction RMS values; closed symbols their experimentally obtained values. Solid lines represent simulations with subfilter combustion model, dashed lines represent simulations in which $\bar{\omega}_Y$ is approximated by $\dot{\omega}_Y$. 
FIGURE 5.13: Centerline FA progress variable and centerline FA progress variable RMS values. Upper lines denote centerline FA progress variable from simulations and open symbols denote values reconstructed from measurements. Lower lines denote centerline FA progress variable RMS values and closed symbols their experimentally obtained values. Solid lines represent simulations with subfilter combustion model, dashed lines represent simulations in which $\dot{\omega}_Y$ is approximated by $\dot{\omega}_Y$. The dash-dotted profile represents the time-average of the estimated subfilter variance by equation (5.24).

FIGURE 5.14: Radial profiles of FA progress variable and FA progress variable RMS values at heights $z/D = 15$, $z/D = 30$ and $z/D = 45$. Upper lines denote FA progress variable from simulations; open symbols denote velocity measurements. Lower lines denote FA progress variable RMS values; closed symbols their experimentally obtained values. Solid lines represent simulations with subfilter combustion model, dashed lines represent simulations in which $\overline{\omega}_Y$ is approximated by $\dot{\omega}_Y$. 
temporarily exceeds the quenching limit, Delhaye [Delhaye2009] obtained an improvement of predictions by FGM tables, when a third control variable, $c_3 = Y_{H_2O} - 8Y_{H_2} - 2Y_{OH}$, is added. The use of this additional control variable, and the resulting additional dimension of the FGM table, might improve $Y$-profiles in Sandia
Another possible explanation stems from preferential diffusion effects, which are absent in the simulation due to the unit Lewis number assumption. Since a high numerical resolution is used in the current LES when compared to previous studies, the inclusion of preferential diffusion effects might lead to a better correspondence between numerical results and experimental data. At both the centerline and height $z/D = 30$ it is clearly visible that the discrepancy between LES results and measurements occurs mainly in rich conditions, i.e. $Z > 0.351$. For these conditions $O_2$, having a Lewis number of approximately 1.1, is the deficient species instead of $CH_4$, having a Lewis number close to unity. Since unit Lewis numbers are used in the FGM table, the diffusion coefficient of the deficient species $O_2$ is overestimated in rich conditions. Barlow et al. [Barlow1998] state that spatial averaging over the probe volume, with a spatial resolution of approximately 750 µm, has very limited influence on measured scalar quantities. Spatial averaging effects are only expected to have a non-negligible influence in high-gradient regions close to the inflow plane ($z/D \leq 3$). In section 4.3.3 it has been shown that preferential diffusion effects are visible in measurement data, although not as strong as in FGM tables using a complex molecular diffusion transport model. These preferential diffusion effects have mainly been observed on the rich side of the stoichiometric plane: exactly where the diffusion of the deficient species $O_2$ is overestimated by the use of unit Lewis numbers. Perhaps the overestimated diffusion coefficient of $O_2$ on the rich side of the stoichiometric plane leads to an overestimated progress variable source term in this region, when compared to experimentally obtained data. In future studies, the influence of preferential diffusion effects can be assessed by taking them into account like in previous FGM studies [Swart2010]. Although $\dot{Y}$ are generally overestimated, RMS values match experimentally obtained values quite well.

Figures 5.11 and 5.13 show that time-averaged subfilter variances are substantially smaller than resolved variances. The fact that their absolute values remain very small explains why the use of the PPDF-integrated FGM table yields nearly identical results as the FGM table without subfilter closure. This is consistent with the identical conclusion drawn from figure 5.8 in section 5.5.1.

### 5.5.3 CO and H$_2$ mass fractions

To obtain species mass fraction predictions two methods are available: direct interpolation from the FGM table and solving a transport equation in which the chemical source term is retrieved from the FGM table. These two methods are used for CO and H$_2$ mass fraction predictions, and compared to measurements. For CO, results from Laser Induced Fluorescence (LIF) measurements are used instead of Raman measurements, because the latter is more affected by hydrocarbon fluorescence interferences in rich conditions [Barlow2003].

In figures 5.17 and 5.18 axial and radial profiles of CO mass fraction predictions are shown in mixture fraction space. The first observation is that solving a transport equation instead of using direct interpolation from the FGM table only slightly modifies CO predictions in rich conditions consistently towards measurements. On the other hand, in the transport equation approach $\dot{Y}_{CO}$ is a function of $Z$ and $\dot{Y}$ only, and is therefore decoupled from its own mass fraction. This implies that the conservation of mass is no longer guaranteed, and non-physical (negative) values for the $Y_{CO}$ can be obtained as is shown in figure 5.18. In both Flame D and F this occurs in regions where $\dot{Z} < 0.2$; these regions correspond to the
5.5 Results

**Figure 5.17:** Centerline FA CO mass fraction in mixture fraction space. Solid line represents direct interpolation from FGM table, dashed line represents solved transport equation. Black lines denote results from the laminar FGM table, red lined denote results using the PPDF subfilter model. Open circles denote measurements and solid squares denote a priori results without subfilter combustion model.

**Figure 5.18:** FA CO mass fraction in mixture fraction space at heights \(z/D = 15\), \(z/D = 30\) and \(z/D = 45\). Solid line represents direct interpolation from FGM table, dashed line represents solved transport equation. Black lines denote results from the laminar FGM table, red lined denote results using the PPDF subfilter model. Open circles denote measurements, solid squares denote a priori results without subfilter combustion model and open diamonds denote a priori results with subfilter combustion model.

lean part of the counterflow diffusion flamelets, from which the FGM table is generated, where CO is oxidized to \(\text{CO}_2\). The possibility to obtain negative mass fraction values is an inherent drawback of the use of an additional transport equation in which a source term
Figure 5.19: Centerline FA $\text{H}_2$ mass fraction in mixture fraction space. Solid line represents direct interpolation from FGM table, dashed line represents solved transport equation. Black lines denote results from the laminar FGM table, red lined denote results using the PPDF subfilter model. Open circles denote measurements and solid squares denote a priori results without subfilter combustion model.

Figure 5.20: FA $\text{H}_2$ mass fraction in mixture fraction space at heights $z/D = 15$, $z/D = 30$ and $z/D = 45$. Solid line represents direct interpolation from FGM table, dashed line represents solved transport equation. Black lines denote results from the laminar FGM table, red lined denote results using the PPDF subfilter model. Open circles denote measurements, solid squares denote a priori results without subfilter combustion model and open diamonds denote a priori results with subfilter combustion model.

From a chemistry table is used. The second observation is that the use of the PPDF subfilter combustion model consistently deteriorates $Y_{\text{CO}}$ predictions, both when direct
interpolation is used or when a transport equation for CO is solved. This might be due to an overestimation of the subfilter variance, since a factor $\alpha = 1/8$ is used in equation (5.24) instead of the theoretical $\alpha = 1/12$ value. The large errors might also originate from the underestimated variances in $Z$ and $Y$ in the region near the nozzle: this underestimation causes high chemical production terms near the nozzle, while the non-negligible variances in the downstream (lean) region reduce the chemical consumption contribution. This might explain the significant overestimation of $Y_{\text{CO}}$ in lean conditions when the PPDF subfilter combustion model is used in conjunction with the use of a transport equation. It might also be that the presumed shape for the PDF does not accurately represent the PDF that would be obtained experimentally. The Conditional Momentum Closure (CMC) approach [Bilger1993, Klimenko1990] might improve results because of a better representation of the experimentally obtained PDF. The CMC approach can be interpreted as a multi-surface model, since a conditional equation is solved for a large number of $Z$-level. CMC is an expensive approach, even though the cost is often decreased by using a coarser numerical grid for the CMC equations than for the CFD equations.

For Sandia Flame D simulation results without the subfilter PPDF combustion model match experimental data quite well, except at height $z/D = 15$. In Flame F, results from the simulations are less satisfactory, especially in rich conditions. When the errors in $Z$ and $Y$ are excluded by performing an a priori analysis using time-averaged $Z$- and $Y$-values, it is seen that predictions are much better than predictions from the numerical simulations. In Flame F, for which numerical results do not very well match experimental data for $z/D < 45$, this was to be expected since figure 5.15 shows that the LES severely overestimate $Y$ for $Z > 0.4$. Furthermore, in the region around the stoichiometric plane ($0.2 < \bar{Z} < 0.4$) taking into account variances of $Z$ and $Y$ further improves a priori results. This implies that accounting for the non-linear response of the CO mass fraction with respect to $Z$ and $Y$ fluctuations improves predictions.

Figures 5.19 and 5.20 show that results for $H_2$ are very similar to those of CO. For this species as well, the main error origin in the numerical simulations are the errors in the $Z$- and $Y$-fields. When the PPDF subfilter combustion model is omitted, predictions tend to be slightly improved in rich conditions, like for CO. However, similar negative $Y_{H_2}$ are obtained in lean regions. For $H_2$, an intensified dissimilarity between the omittance and use of the subfilter combustion model is observed when compared to results for CO. The use of the subfilter combustion model results in severe overestimation of $Y_{H_2}$ in rich conditions. The use of a transport equation for $H_2$ in conjunction with the PPDF subfilter combustion model does not improve results, the overestimation is merely shifted to leaner conditions. When these errors are excluded in an a priori analysis, mass fraction values from the FGM table nearly coincide with experimentally obtained values. It is remarked that the accounting for $Z$ and $Y$ variances does not yield a significant improvement in a priori $H_2$ mass fraction predictions. This might be explained by the less smooth surface representing the CO mass fraction in the FGM table, than the surface that is found for $H_2$. Both surfaces are depicted in figure 5.21.

5.6 Conclusions

In this chapter, predictions for CO and $H_2$ in Large Eddy Simulations (LES) of partially-premixed turbulent flames have been compared with measurements. In chapter 4 it was concluded that FGM tables can yield accurate predictions for species mass fractions,
even for notorious species like CO and H₂, provided that applicable modeling
assumptions are made when constructing the FGM table. In this chapter, the error which
originates from inaccuracies in control variable values in LES, is added to the errors which
stem from chemistry tabulation, discussed in section 4.2. The best-performing FGM table
from the \emph{a priori} analysis in chapter 4 is used in the simulations of Sandia Flame D and F.

The flow field, and as a consequence the field of passive scalar \( Z \), are not very well
recovered in the current simulations. This is attributed to the prescribed turbulent inflow
field, which is a poor representation of the inflow conditions in the experiments. However,
when \( Y', Y_{CO} \)- and \( Y_{H2} \)-results from simulations are depicted as a function of \( Z \) instead of
spatial coordinate, it is observed that for Flame D the flame structure is fairly well
recovered. For Flame F, which exhibits significantly more non-equilibrium effects, the
flame structure is less satisfactory recovered due to an overestimation of the progress
variable source term in rich conditions. For both flames the inclusion of a Presumed PDF
subfilter combustion model, using a \( \beta \)-PDF in both \( Z \)- and \( Y' \)-direction, does not
significantly influence numerical result. Since the eddy-viscosity is dominated by the
molecular viscosity in the region where the flame brush crosses the centerline, \( z/D \approx 30 \)
and \( z/D \lesssim 45 \), it is expected that the used turbulent Schmidt number, \( ScT = 0.6 \) in this
study compared to \( ScT = 0.4 \) in other studies [Pitsch2000, Vreman2008], and the related
lowered eddy-diffusivity does not significantly influence numerical outcomes.

Predictions for \( Y_{CO} \) and \( Y_{H2} \) slightly improve in rich conditions when a transport equation,
using the species its source term from the FGM table, is solved instead of directly
interpolation from the FGM table. However, this approach no longer guarantees mass
conservation and might lead to non-physical mass fraction predictions as observed in
current simulations for both CO and H₂ in lean conditions. In the current implementation
the chemical source term for these species does not depend on the species its mass fraction
itself, \( i.e. \) no direct coupling between the species mass fractions and their chemical source
term exists. The addition of a species mass fraction as a control variable, \( \dot{\omega}_{CO} = \dot{\omega}_{CO} (Z, Y, Y_{CO}) \) instead of \( \dot{\omega}_{CO} = \dot{\omega}_{CO} (Z, Y) \), might improve results because it
links the species source term to its own mass fraction.

When errors in control variables \( Z \) and \( Y' \) are excluded by means of an \emph{a priori} analysis, is
is found that results match experimental data much better than LES results. The relatively
smooth behavior of H₂ in the FGM table makes accounting for variances of \( Z \) and \( Y' \) in the
\emph{a priori} analysis superfluous; for accurate \emph{a priori} results for CO these variances should be
taken into account. Like in chapter 4.2, it is concluded that an FGM table can yield

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig521.png}
\caption{CO and H₂ mass fraction as a function of Z and C in the FGM table. Only
every fifth entry for Z and tenth entry for C is shown.}
\end{figure}
accurate predictions for these species, provided that $Z$ and $Y$ closely resemble those observed in experiments. In order to obtain accurate $Z$ and $Y$ fields, a very important prerequisite is an accurate prescription of turbulent inflow boundary conditions, which greatly influences passive scalar fields.
Combined FGM approach

6.1 Introduction

The Flamelet Generated Manifold (FGM) [Oijen2000] reduction method is based on the assumption that trajectories in composition space in multi-dimensional flames closely resemble trajectories of laminar, one-dimensional flames denoted by the name flamelet [Peters1984]. In this reduction method a low-dimensional chemical manifold is generated from a (large) number of flamelets, and can therefore be considered as a combination of classic flamelet- and manifold methods. During a flame simulation, only transport equations for the controlling variables have to be solved: all dependent thermochemical variables are retrieved from the FGM table. Originally, the FGM reduction method was based on premixed flamelets but in general other flamelet types can be used just as well. In chapters 4 was shown that for highly-stratified partially-premixed flames, the Sandia Flames, an FGM table based on counterflow diffusion flamelets yield much better species mass fraction predictions than an FGM table based on premixed flamelets. Subsequently, the most applicable FGM table, generated from (partially-premixed) counterflow diffusion flamelets, has been applied in Large Eddy Simulations (LES) simulations of Sandia Flame D and F in chapter 5. For these flames it is well-known that they exhibit a quasi non-premixed character, although the fuel stream contains 75 % of air by volume. The choice which type of flamelets to use for the generation of an FGM table, then is a very straightforward one. This question becomes more difficult to answer for flames in which premixing in the reactor becomes more important.

FGM tables composed of steady premixed flamelets have proven to be very accurate for Bunsen-type flames including heat loss effects [Oijen2000], highly stretched premixed counterflow flames [Oijen2002-1] and confined triple flames [Oijen2004]. Partially-premixed counterflow flames with a fuel equivalence ratio up to two have also been shown to be able to be accurately described by premixed flamelet tables [Bongers2005-2, Fiorina2005-1] but in both simulations of laminar and turbulent [Bradley2002, Vervisch2004, Vreman2008] flames with higher fuel equivalence ratios significant inaccuracies were observed for typical reaction product species in rich conditions like H$_2$ and CO. Inaccuracies can be attributed, besides inexactness of the flowfield representation, to inaccurate predictions of controlling variables, which causes wrong look-up locations in the chemistry table, and the fact that premixed flamelet-based tables do not allow diffusion in mixture fraction direction. Chapter 4 showed that even with exact values for the controlling variables, a premixed flamelet-based FGM can not yield satisfactory predictions for H$_2$ and CO in fuel rich regions. For the Sandia Flames discussed in chapter 4, a counterflow diffusion flamelet-based FGM table using the observed minimum and maximum equivalence ratios as boundary conditions obtained far better results. However, in many practical applications degree of premixing inside a
combustion chamber is not exactly known, making it impossible to generate a dedicated
FGM table based on partially-premixed counterflow flamelets with the correct boundary
conditions.

For partially premixed applications an online method is desired, which can distinguish
between a premixed and non-premixed character locally; thermochemical data from the
most appropriate chemistry table is then retrieved and used by the CFD solver. Fiorina
et al. [Fiorina2005-1] introduced a modified version of the flame-index of Yamashita
et al. [Yamashita1996] to combine the progress variable chemical source term from a
(premixed flamelet-based) FPI database and a steady strained non-premixed flamelet in
mixture fraction space [Peters2000], i.e.

\[
\dot{\omega}_i = -\rho \chi_z \left( \frac{\partial^2 Y_i}{\partial Z^2} \right)_\text{FPI},
\]

in which \( Y_i \) and \( \dot{\omega}_i \) denote a species its mass fraction and chemical source. \( \rho \), \( Z \) and \( \chi_z \) denote density, the mixture fraction and the scalar dissipation rate respectively. In other
words, no database constructed from diffusion flamelets was used for the non-premixed
combustion mode and in the source term approximation, equation (6.1), species gradients
in mixture fraction space from a premixed flamelet-based table have been used. Since a
transport equation for the progress variable was solved, the discrete transition between the
source terms from the FPI table and the steady strained non-premixed flamelet solution is
smoothed out.

In this chapter, a novel approach to combine premixed and counterflow diffusion
flamelet-based FGM tables is proposed for representation of combustion chemistry in
partially-premixed flames. A non-discrete switch function will be introduced, which
combines values for thermochemical variables from premixed and counterflow diffusion
flamelet-based FGM tables using a weighting function which depends on local conditions.
In contrast to previously proposed (discrete) switches [Yamashita1996, Fiorina2005-1], this
switch-function ensures a smooth transition from the premixed flamelet-based FGM to the
non-premixed flamelet-based FGM and vice versa. It is assessed whether the combined
use of premixed and counterflow diffusion flamelet-based FGM tables can be used to
obtain accurate \( \text{H}_2 \) and \( \text{CO} \) mass fraction predictions. This would eliminate having to \textit{a priori} choose one flamelet type for the generation of FGM tables.

**Chapter structure**

In section 6.2 the governing equations for both premixed and counterflow diffusion
flamelets are given and the generation procedure for FGM databases is explained briefly.
Subsequently, in chapter 6.3 it is explained how FGM tables can replace detailed chemistry
and that the type of flamelet on which the FGM is based, has a decisive influence on
simulation results. Section 6.4 explains the weighting procedure of premixed and
counterflow diffusion flamelet-based FGM tables and validates this approach for the two
limits of a broad spectrum of partially-premixed flamelet: steady premixed flamelets and
steady non-premixed counterflow diffusion flamelets. Subsequently, for
partially-premixed flamelets with different stratification levels and applied strain rates
results in terms of flame structure and mass fractions of \( \text{OH}, \text{H}_2 \) and \( \text{CO} \) are analyzed.
Vreman et al. [Vreman2008] showed in LES of Sandia Flame D that a much better
prediction for \( \text{NO} \) can be obtained when an additional transport equation for this species,
in which the chemical source term from the FGM table is used, is solved instead of direct retrieval of the mass fraction from the FGM table. Section 6.6 examines whether this approach can result in an improvement of predictions for the notoriously slow-evolving CO mass fraction. Finally, summarizing conclusions are given in section 6.7.

6.2 Flamelet equations and FGM generation

The flamelet equations form a set of specific transport equations for one-dimensional reacting flows describing conservation of mass, species and enthalpy. Two specific types will be used to generate FGM tables: premixed freely-propagating flamelets and non-premixed counterflow diffusion flamelets. In this section, for both type the governing equations are discussed briefly and subsequently the FGM generation procedure is explained.

Methane-air flames are treated in this chapter: the assumption of unit Lewis numbers is appropriate to these fuel-air mixtures which allows the simplification of the transport equations. Law and Sung [Law2000] state that the Lewis number of the deficient species, *i.e.* CH$_4$ in lean mixtures and O$_2$ in rich mixtures, is the mixture’s effective Lewis number. The Lewis number of CH$_4$ is very close to unity and the Lewis number of O$_2$ approximately equals 1.1. Because both Lewis numbers are close to unity, it is assumed that the unit Lewis number assumption is appropriate for the flames discussed in this chapter.

**Flamelet equations**

The equations describing steady, stretchless premixed flamelets have been derived in section 2.2.1 and read:

\[
\frac{\partial \rho u}{\partial x} = 0 , \tag{6.2}
\]

\[
\frac{\partial \rho u Y_i}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda}{L_{ei} c_p} \frac{\partial Y_i}{\partial x} \right) + \dot{\omega}_i, \quad i \in [1, N_s - 1] \tag{6.3}
\]

\[
\frac{\partial \rho u h}{\partial x} = \frac{\partial}{\partial x} \left[ \frac{\lambda}{c_p} \frac{\partial h}{\partial x} + \frac{\lambda}{c_p} \sum_{i=1}^{N_s} \left( \frac{1}{L_{ei}} - 1 \right) h_i \frac{\partial Y_i}{\partial x} \right] , \tag{6.4}
\]

\[
\rho = p_\infty M / (RT) , \tag{6.5}
\]

in which the Low Mach-number approximation is used in the thermal equation of state (6.5) with $p_\infty$ denoting the ambient pressure. Dirichlet boundary conditions for $Y_i$ and $h$ are imposed at the unburnt side while Neumann boundary conditions for these variables are imposed at the chemical equilibrium side:

\[
Y_i (x \to -\infty) = Y_{i,-\infty} , \quad h (x \to -\infty) = h_{-\infty} , \tag{6.6}
\]

\[
\frac{\partial Y_i}{\partial x} (x \to \infty) = 0 , \quad \frac{\partial h}{\partial x} (x \to \infty) = 0 . \tag{6.7}
\]

The translational degree of freedom is removed by choosing a fixed temperature at $x = 0$ and flamelet solutions are fully defined by their boundary conditions at $x \to -\infty$. 

The equations describing quasi-steady, counterflow diffusion flamelets were derived in section 2.2.2:

\[
\frac{\partial \rho}{\partial t} + \frac{\partial \rho u}{\partial x} = -\rho G ,
\]
(6.8)

\[
\frac{\partial \rho Y_i}{\partial t} + \frac{\partial \rho u Y_i}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda}{Le_i c_p} \frac{\partial Y_i}{\partial x} \right) + \dot{\omega}_i - \rho Y_i G , \quad i \in [1, N_s - 1] ,
\]
(6.9)

\[
\frac{\partial \rho h}{\partial t} + \frac{\partial \rho u h}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda}{c_p} \frac{\partial h}{\partial x} \right) + \frac{\lambda}{c_p} \sum_{i=1}^{N_s} \left( \frac{1}{Le_i} - 1 \right) h_i \frac{\partial Y_i}{\partial x} - \rho h G ,
\]
(6.10)

\[
\frac{\partial \rho G}{\partial t} + \rho u \frac{\partial G}{\partial s} = \frac{\partial}{\partial s} \left( \mu \frac{\partial G}{\partial s} \right) + \rho_{ox} \left( \frac{\partial a(t)}{\partial t} + a(t)^2 \right) - \rho G^2 .
\]
(6.11)

in which strain rate \( G \) is defined as \( G \equiv \frac{\partial v}{\partial y} \). The derivation of equation (6.11) can be found in appendix A. The unsteady formulation allows the prescription of the applied strain rate as a function of time. Contrary to steady flamelet solutions, which can not be obtained beyond the extinction strain rate, unsteady flamelets can be tracked from an initial low strain rate to the mixing solution, thereby guaranteeing a smooth continuation at the extinction limit. The Low Mach-number approximation for the equation of state, equation (6.5), is used to close the set of equations. Dirichlet boundary conditions for \( Y_i, h \) and \( G \) are imposed in the oxidizer \( (x \to -\infty) \) stream while in the fuel \( (x \to \infty) \) stream a Neumann boundary condition for \( G \) is imposed:

\[
Y_i (x \to \infty) = Y_{i,ox} , \quad h (x \to \infty) = h_{ox} , \quad G (x \to \infty) = a(t) ,
\]
(6.12)

\[
Y_i (x \to -\infty) = Y_{i,fu} , \quad h (x \to -\infty) = h_{fu} , \quad \frac{\partial G}{\partial x} (x \to -\infty) = 0 .
\]
(6.13)

The applied strain rate \( a(t) \) which is defined at the oxidizer side \( (x = \infty) \), together with fuel and oxidizer composition and temperature completely defines the flamelet solution. The translational degree of freedom is removed by choosing the stagnation plane at \( x = 0 \).

For both the premixed and the counterflow flamelets, simplified formulations for \( \mu \) and \( \lambda \) for CH\(_4\)-air flames are used [Smooke1991] to reduce computational cost:

\[
\mu_{L/c_p} = 1.67 \times 10^{-8} \left( T/298 \right)^{0.51} ,
\]
(6.14)

\[
\lambda/c_p = 2.58 \times 10^{-5} \left( T/298 \right)^{0.69} .
\]
(6.15)

Source terms for species are computed using the GRI 3.0 reaction scheme [GRI mech30]; the mass fraction of the abundant species N\(_2\) is determined using the identity \( \sum Y_i = 1 \). All flamelet computations have been performed with the in-house flame code Chem1D [Somers1994] and in the premixed flamelets the chemical equilibrium is explicitly added to each flamelet to ensure the correct equilibrium composition for slow evolving species. Due to the limited domain size, chemical equilibrium might not be fully reached inside the numerical domain for small or large values of \( \phi \). Adding the chemical equilibrium state explicitly prevents the omission of this state in the FGM table.

**FGM parametrization and generation**

Flamelet solutions are mapped on two control variables: mixture fraction \( Z \) describing mixing and reaction progress variable \( Y \) describing reaction progress, viz.

\[
\varphi = \varphi (Z, Y) ,
\]
(6.16)
in which $\varphi$ can denote any thermochemical variable. $Z$ is a normalized scalar based on element composition [Bilger1990]:

$$
Z = \frac{2 M_H^{-1} [Z_H - Z_{H,2}] + 0.5 M_C^{-1} [Z_C - Z_{C,2}] - M_O^{-1} [Z_O - Z_{O,2}]}{2 M_H^{-1} [Z_{H,1} - Z_{H,2}] + 0.5 M_C^{-1} [Z_{C,1} - Z_{C,2}] - M_O^{-1} [Z_{O,1} - Z_{O,2}]}, \quad (6.17)
$$

in which $Z_k$ denotes the molar mass of an element $k$ and subscripts $H$, $C$, $O$, 1 and 2 refer to hydrogen, carbon, oxygen and the fuel- and oxidizer stream, respectively. The equivalence ratio $\varphi$ relates to the mixture fraction by

$$
\varphi(Z) = \frac{s Y_{F,1}}{Y_{O_{2,2}} (\frac{Z}{1 - Z})}, \quad (6.18)
$$

in which $s$ denotes the stoichiometric oxidizer/fuel mass ratio, $Y_{F,1}$ denotes the fuel mass fraction in the fuel stream and $Y_{O_{2,2}}$ denotes the $O_2$ mass fraction in the oxidizer stream ($s = 3.989$ for $CH_4$-air flames; $Y_{O_{2,2}} = 0.233$ for air). For the reaction progress variable $Y$ a linear combination of species mass fractions

$$
Y = \sum_{i=1}^{N_s} \alpha_i Y_i, \quad (6.19)
$$

in which $\alpha_i = 0$ for all species except for $CO_2$, $H_2O$ and $H_2$ is applied. Weight factors for these species are $\alpha_{CO_2} = M_{CO_2}^{-1}$, $\alpha_{H_2O} = M_{H_2O}^{-1}$ and $\alpha_{H_2} = M_{H_2}^{-1}$ which ensures unambiguous mapping of methane-air flames between its lower ($\varphi = 0.35$) and upper ($\varphi = 3.6$) flammability limit. For the definition of $Y$ given by equation (6.19), $Y_0 = 0$ for any given value of $Z$.

For the FGM table based on steady, stretchless premixed flamelets, henceforth indicated by P-FGM, approximately 425 flamelet solutions have been computed for equidistantly-spaced values of $Z$ between the flammability limits. Beyond flammability limits, thermochemical variables have been computed by linear interpolation between the oxidizer stream and lower flammability limit or upper flammability limit and fuel stream. Species mass fractions and temperature are interpolated on an equidistant $Z$-$Y$ grid and subsequently $\rho$ and $c_p$ are recalculated on the (equidistant) $Z$-$Y$ grid.

For the FGM table based on counterflow diffusion flamelets, henceforth indicated by NP-FGM, a non-premixed flamelet with a very low applied strain rate, i.e. close to chemical equilibrium, is used. By increasing the applied strain rate as a function of time, the flamelet is pushed towards its extinction limit. When extinction is achieved, flamelet solutions are tracked in time all the way to the mixing limit; this method ensures a smooth continuation at the extinction limit. Subsequent flamelet solutions in time with a monotonously decreasing $Y$ are used to construct the NP-FGM. In figure 6.1 the progress variable source term from a P-FGM and the difference in progress variable $\Delta \dot{\omega}_Y = (\dot{\omega}_Y^{NP} - \dot{\omega}_Y^{P})$ between a NP-FGM and a P-FGM are depicted. In this figure it can be seen that that $\Delta \dot{\omega}_Y$ has the same order of magnitude as $\dot{\omega}_Y$; for other thermochemical variables similar differences between the two tables are found. The shape of the right graph in figure 6.1 indicates that the maximum $\dot{\omega}_Y$-values in the counterflow diffusion flamelet-based FGM table are found at higher $Z$-values than in the premixed flamelet-based table.
FIGURE 6.1: Progress variable source \( \dot{\omega}_Y \) retrieved from a premixed flamelet-based FGM (left) and the difference in progress variable source \( \Delta \dot{\omega}_Y = \dot{\omega}_Y^{NP} - \dot{\omega}_Y^P \). \( C \) denotes the normalized progress variable \( C = \frac{Y}{Y_{eq}} \). Only the region of significance has been shown; in \( C \)-direction only every eight database entry is shown.

6.3 Application of FGM tables in one-dimensional flames

In simulations of flamelets now either detailed chemistry, involving solving a transport equation for each species, or FGM chemistry can be used. In the latter case only a differential equation for controlling variables \( Z \) and \( \mathcal{Y} \), in combination with conservation equations for mass and momentum, has to be solved and dependent thermochemical variables are retrieved from an FGM table. One-dimensional transport equations for \( Z \) and \( \mathcal{Y} \) in flamelets read:

\[
\frac{\partial \rho Z}{\partial t} + \frac{\partial \rho u Z}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda}{c_p} \frac{\partial Z}{\partial x} \right) - \rho Z G , \tag{6.20}
\]

\[
\frac{\partial \rho \mathcal{Y}}{\partial t} + \frac{\partial \rho u \mathcal{Y}}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda}{c_p} \frac{\partial \mathcal{Y}}{\partial x} \right) + \dot{\omega}_Y - \rho \mathcal{Y} G , \tag{6.21}
\]

for which \( \rho, \dot{\omega}_Y \) and \( T \) are retrieved from the FGM table during a computation; the latter is required for the evaluation of equations (6.14-6.15). For premixed flamelets with unit Lewis numbers, i.e. without preferential diffusion, \( Z \) is a constant value implying that equation (6.20) can be removed from the system of equations. For steady, stretchless premixed flamelets, the time-dependent term and the stretch term drop out of equation (6.21) as well.

In a premixed flamelet with a given \( Z \)-value, a Dirichlet boundary condition for \( \mathcal{Y} \) is imposed at the unburnt side and a Neumann boundary conditions at the chemical equilibrium side:

\[
\mathcal{Y} (x \to -\infty) = \sum_{i=1}^{N_s} \alpha_i Y_i (x \to -\infty) , \quad \frac{\partial \mathcal{Y}}{\partial x} (x \to \infty) = 0 , \tag{6.22}
\]

in which \( \alpha_i \) was defined in section 6.2. Inversely, for a given value of \( Z \), thermochemical variable values in the unburnt mixture can be obtained by:

\[
\varphi = Z \varphi_{fu} + (1 - Z) \varphi_{ox} , \tag{6.23}
\]

where \( \varphi \) can denote \( Y_i \) or \( h \). For a counterflow diffusion flamelet, Dirichlet boundary conditions for \( Z \) are imposed in the fuel and oxidizer stream and boundary conditions for
6.3 Application of FGM tables in one-dimensional flames

\[ \mathcal{Y} \] follow from the composition of fuel and oxidizer and equation (6.19):

\[ Z(x \to -\infty) = 1, \quad Z(x \to -\infty) = 0, \]

\[ \mathcal{Y}(x \to -\infty) = \sum_{i=1}^{N_s} \alpha_i Y_{i,\text{fu}}, \quad \mathcal{Y}(x \to \infty) = \sum_{i=1}^{N_s} \alpha_i Y_{i,\text{ox}}. \]  

(6.24)

(6.25)

For one-dimensional flames addressed in this chapter, pure fuel consists of CH\textsubscript{4} only and the oxidizer is air \((Y_{O_2,\text{air}} = 0.233; Y_{N_2,\text{air}} = 0.767)\). These compositions imply that for premixed flames \(\mathcal{Y}(x \to -\infty) = 0\) and for counterflow diffusion flames \(\mathcal{Y}(x \to -\infty) = \mathcal{Y}(x \to \infty) = 0\) since CO\textsubscript{2}, H\textsubscript{2}O and H\textsubscript{2} are absent in both fuel and oxidizer.

An FGM table can be generated with a user-defined resolution in all control variable directions. In this study an equidistant spacing has been used, both in Z and Y-direction. Data is retrieved from the table by bi-linear interpolation, hence an interpolation error proportional to the square of the FGM table its resolution is introduced. The influence of the interpolation error is illustrated by figure 6.2 in which the global error \(\varepsilon_{\mathcal{Y}}\) is defined as

\[ \varepsilon_{\mathcal{Y}} = \int_0^Z (\mathcal{Y}^{\text{FGM}} - \mathcal{Y}^{\text{DET}}) dZ \left[ \int_0^1 (\mathcal{Y}^{\text{DET}}) dZ \right]^{-1}. \]

(6.26)

The left graph of figure 6.2 depicts two one-dimensional flames for which either a 21 \(\times\) 21 or a 401 \(\times\) 401 FGM table is used: it is clearly visible that the more refined table yields a much more accurate result. In the right part of figure 6.2 can be seen that the slope of the exponential fit through global error values, which equals \(-1.72\), corresponds quite well to the expected value of \(-2\). In this study the maximum table size of 401 \(\times\) 401 \((Z \times Y)\) will be used; this results in a table size of approximately 225 MB when all species from the GRI 3.0 reaction mechanism [GRImech30] are tabulated.

**Appropriate flamelet type for applications**

The necessity for the use of the appropriate type of flamelet to generate an FGM table becomes apparent when one-dimensional premixed and non-premixed flames are computed using both a P-FGM and a NP-FGM. Premixed CH\textsubscript{4}-air (21% O\textsubscript{2} and 79% N\textsubscript{2} by volume) flames with equivalence ratios of 0.67, 1.00 and 1.50 are depicted in the left part of

---

**Figure 6.2:** Structure (left) and global error (right) for \(\mathcal{Y}\) in a non-premixed one-dimensional flame with an applied strain rate of 100 s\(^{-1}\) for different FGM resolutions. In the left graph, the flame structure obtained using a 21 \(\times\) 21 \((Z \times Y)\) table (dashed line) and a 401 \(\times\) 401 table (solid line) are shown in Z-space. The right graph depicts global error, defined by equation 6.26, for multiple table resolutions (symbols) and the fit of an exponential function (solid line) as a function of the number of entries in the FGM table.
Combined FGM approach

Figure 6.3. Similar to the premixed flames, non-premixed flames with strain rates of 20, 100 and 500 [1/s] are depicted in the right part of figure 6.3; the stoichiometric mixture fraction equals 0.055. Obtained solutions of equations (6.2-6.4) for premixed flames or equations (6.8-6.11) for non-premixed flames, using the GRI 3.0 reaction mechanism [GRImech30] for the chemical source terms, are compared with solutions of equations (6.20-6.21) using FGM tabulated chemistry for the progress variable source term. The P-FGM and NP-FGM have been generated in a pre-processing step for identical conditions, i.e. atmospheric pressure \( p_0 = 1.01325 \times 10^5 \) Pa and an initial temperature of 300 K for all unburnt gas streams (gas mixture in premixed case; both fuel and oxidizer stream in non-premixed case). In the left part of figure 6.3 it can be seen that the flame thickness of premixed flames, and thereby the laminar flame propagation velocity \( s_L \), is relatively well predicted by the NP-FGM for \( \phi = 0.67 \) and \( \phi = 1.00 \) but is severely underestimated for \( \phi = 1.50 \). The \( s_L \)-based error, defined as

\[
\epsilon = \left| \frac{s_{fgm}^L - s_L}{s_L} \right| ,
\]

remained less than 0.003 for all three cases when the P-FGM is used as is to be expected. In definition (6.27) \( s_L \) denotes the flame propagation velocity which is obtained using detailed chemistry and \( s_{fgm}^L \) denotes the flame propagation velocity which is obtained when an FGM table is used to represent combustion chemistry. Numerical values for the predicted flame thicknesses and errors can be found in table 6.1. The progress variable profile in non-premixed flamelets was increasingly underestimated with increasing applied strain rate as can be seen in the right part of figure 6.3. For small applied strain rate values, counterflow diffusion flame solutions produced by either FGM table are close to equilibrium; the values for the progress variable source term predicted by the FGM tables are close to each other. For higher applied strain rate values, the P-FGM based progress variable profile deviates from the detailed chemistry solution. The underestimation of the progress variable source term by the P-FGM table in rich conditions causes the deviation from the NP-FGM (and the detailed) solution for rich mixture fraction values. Inversely, the large error in flame thickness prediction for the rich (\( \phi = 1.50 \)) premixed flame is caused by the overestimation of the progress variable source term by the NP-FGM. For the non-premixed flame an error is based on the maximum temperature:

\[
\epsilon = \left| \frac{T_{fgm}^{max} - T_{max}}{T_{max}} \right| ,
\]

in which \( T_{max} \) denotes the maximum temperature which is obtained using detailed chemistry and \( T_{fgm}^{max} \) denotes the maximum temperature which is obtained when an FGM table is used to represent combustion chemistry. From table 6.1 can be seen that errors in \( T_{max} \) are two orders of magnitude smaller when an NP-FGM is used instead of a P-FGM.

In the left part of figure 6.4 it can be seen that CO mass fraction values are very well predicted by the P-FGM for premixed flames. The NP-FGM severely overestimates CO mass fraction values in the lean (\( \phi = 0.67 \)) premixed flame because diffusion is allowed across mixture fraction values in the NP-FGM table; CO produced in rich regions can diffuse towards lean regions. The inverse occurs in the rich (\( \phi = 1.50 \)) premixed flame where the produced CO is allowed to diffuse towards the stoichiometric and lean regions where it is consumed. The P-FGM does not allow diffusion in mixture fraction direction. In the right part of figure 6.4 it can be seen that the P-FGM overestimates the CO mass fraction in the rich part of non-premixed flames. This error is slightly offset by the underestimated progress variable at high strain rates: if the correct \( Z-Y \) profiles were
Figure 6.3: Spatial structure of progress variable in premixed flamelets with equivalence ratios of 0.66, 1.00 and 1.50 (left) and progress variable in mixture fraction space in non-premixed flamelets with applied strain rates of 20, 100 and 500 s\(^{-1}\) (right). Circles indicate the detailed solution, solid lines indicate results obtained using a P-FGM and dashed lines indicate results obtained using a NP-FGM. The vertical dashed line at \(Z = 0.2\) in figures on the right indicates the scale change of the horizontal axis.

Taken from the detailed simulation, the CO mass fraction would be even more overestimated since CO mass fraction increases with increasing \(\phi\) in the P-FGM in this region. At \(Z = 0.175\) a discontinuity can be observed in the CO mass fraction profile predicted by the P-FGM which is caused by the linear interpolation of species beyond this upper flammability limit for premixed flamelets. For \(H_2\), results similar to those for CO are obtained (not shown).

It can be concluded that for accurate predictions of \(\phi\) and species mass fractions it is of key importance that the appropriate FGM table is used. In the next section it will be examined whether partially-premixed flames can effectively be described by combining P-FGM and NP-FGM tables based on local parameters. The objective is to do so without the use of additional controlling variables, which would result in FGM tables of an impractical (large) size.
Combined FGM approach

<table>
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<tr>
<th></th>
<th>(s_L)</th>
<th>(s_{L,p})</th>
<th>(e^p)</th>
<th>(s_{L,\text{np}})</th>
<th>(e^{\text{np}})</th>
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<tr>
<td>(\phi = 0.67)</td>
<td>16.30</td>
<td>16.31</td>
<td>6.32·10^{-4}</td>
<td>11.71</td>
<td>2.82·10^{-1}</td>
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<tr>
<td>(\phi = 1.00)</td>
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<td>27.96</td>
<td>9.93·10^{-4}</td>
<td>22.68</td>
<td>1.90·10^{-1}</td>
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<td>7.658</td>
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<th>(T_{\text{p, max}})</th>
<th>(e^p)</th>
<th>(T_{\text{np, max}})</th>
<th>(e^{\text{np}})</th>
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<tr>
<td>(a = 20)</td>
<td>2081.4</td>
<td>2036.9</td>
<td>2.14·10^{-2}</td>
<td>2083.4</td>
<td>9.44·10^{-4}</td>
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<tr>
<td>(a = 100)</td>
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<td>1953.3</td>
<td>3.73·10^{-2}</td>
<td>2029.4</td>
<td>2.35·10^{-4}</td>
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<tr>
<td>(a = 500)</td>
<td>1889.3</td>
<td>1792.3</td>
<td>5.14·10^{-2}</td>
<td>1889.1</td>
<td>9.85·10^{-5}</td>
</tr>
</tbody>
</table>

Table 6.1: Premixed part: laminar flame propagation velocities in cm/s for premixed flames for \(\phi = 0.67\), \(\phi = 1.00\) and \(\phi = 1.50\) (depicted in the left part in figure 6.3) predicted by detailed chemistry \((s_L)\), a P-FGM \((s_{L,p})\) and a NP-FGM \((s_{L,\text{np}})\). Errors in \(s_L\) are defined by equation (6.27). Non-premixed part: maximum temperatures for non-premixed flames with applied strain rates of 20, 100 and 500 \(\text{s}^{-1}\) (depicted in the right part in figure 6.3) predicted by detailed chemistry \((T)\), a P-FGM \((T_{\text{p}})\) and a NP-FGM \((T_{\text{np}})\). Errors in \(T\) are defined by equation (6.28).

### 6.4 Combination of FGM tables

Fiorina et al. [Fiorina2005-1] were one of the first to propose a switching strategy to distinguish a premixed and non-premixed burning mode and to use the most applicable chemistry table. Their switching strategy performed quite well in distinguishing the burning modes. Unfortunately, the binary switch they used caused discontinuities in species profiles. The current strategy to combine P-FGM and NP-FGM tables identifies the mixture fraction dissipation rate \(\chi_Z\) and a analogous (normalized) progress variable dissipation rate \(\chi_Y\) as indicators of the local burning mode. For unit Lewis numbers and the definition for \(Y\) given by equation (6.19) they are defined by

\[
\chi_Z(x; Z(x)) = \frac{2\lambda}{\rho_c \rho_p} \left| \frac{\partial Z}{\partial x} \right|^2,
\]

(6.29)

\[
\chi_Y(x; Z(x), Y(x)) = \frac{2\lambda}{\rho_c \rho_p} \left| \frac{1}{Y_{\text{eq}}(Z)} \frac{\partial Y}{\partial x} \right|^2.
\]

(6.30)

The factor \([Y_{\text{eq}}(Z)]^{-1}\) is included in order to normalize \(\chi_Y\). It is proposed to use the ratio \(\chi_Y/\chi_Z\) to identify which combustion mode is (locally) present. In premixed flames, in the absence of preferential diffusion, \(\chi_Z\) equals zero but in non-premixed flames \(\chi_Y\) does not reduce to zero. It is therefore proposed to subtract the spatial progress variable gradient observed in non-premixed flamelets from the (locally) observed spatial progress variable gradient to ensure that the corrected (normalized) progress variable dissipation rate \(\chi_{Y^*}\), defined by

\[
\chi_{Y^*}(x; Z(x), Y(x)) = \frac{2\lambda}{\rho_c \rho_p} \left| \frac{1}{Y_{\text{eq}}(Z)} \left( \frac{\partial Y}{\partial x} - \frac{\partial Z}{\partial x} \frac{\partial Y}{\partial Z} \right)_{\text{NP-FGM}} \right|^2,
\]

(6.31)

reduces to zero when a non-premixed flame is considered. For premixed flames \(\chi_{Y^*}/\chi_Z \rightarrow \infty\) while for non-premixed flames \(\chi_{Y^*}/\chi_Z \rightarrow 0\).

It is assumed that a flame exhibits a premixed character when \(\chi_{Y^*} \gg \chi_Z\) and, inversely, exhibits a non-premixed character when \(\chi_{Y^*} \ll \chi_Z\). To define a switch strategy based on
the $\chi_{Yc}/\chi_{Z}$ ratio, approximately 1000 partially-premixed flamelets are computed by varying applied strain rate ($a \in [20, 500]$ [1/s]) and stratification level ($\Delta \Phi = \Phi_{fu} - \Phi_{ox} \in [0, 1]$) where $\Phi$ is defined as a normalized equivalence ratio:

$$\Phi = \phi / (1 + \phi) .$$

(6.32)

All flamelets have symmetric boundary conditions in $\Phi$: $\Phi = 0.5(\Phi_{fu} + \Phi_{ox}) = 0.5$. In the left part of figure 6.5 spatial mixture fraction profiles are given for one-dimensional counterflow flames with applied strain rates equal to $a = 100$ [1/s]. Figure 6.6 shows the

<table>
<thead>
<tr>
<th>$\alpha_i$</th>
<th>$\omega_{Yc}$</th>
<th>$\rho$</th>
<th>$T$</th>
<th>$Y_{CO}$</th>
<th>$Y_{H_2}$</th>
</tr>
</thead>
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<tr>
<td>$\alpha_1$</td>
<td>$4.82\cdot10^{-1}$</td>
<td>$8.16\cdot10^{-1}$</td>
<td>$3.65\cdot10^{-1}$</td>
<td>$1.98\cdot10^{-1}$</td>
<td>$2.42\cdot10^{-1}$</td>
</tr>
<tr>
<td>$\alpha_2$</td>
<td>$-3.15\cdot10^{-2}$</td>
<td>$-2.51\cdot10^0$</td>
<td>$-6.12\cdot10^{-1}$</td>
<td>$-3.83\cdot10^{-1}$</td>
<td>$-3.40\cdot10^{-1}$</td>
</tr>
</tbody>
</table>

**Table 6.2:** Modeling coefficients $\alpha_i$ as used in equation (6.34) for $\omega_{Yc}$, $T$, $Y_{CO_2}$, $Y_{CO}$, and $Y_{H_2}$.
FIGURE 6.5: Left graph: Spatial mixture fraction profiles for different stratification levels $\Delta \Phi = 0.8$ and $\Delta \Phi = 0.4; \Delta \Phi$ is proportional to $\Delta Z$. All applied strain rates equal $a = 100 \, [1/s]$. Right graph: ratio of dissipation rates $\chi_{Y^*}$ and $\chi_{Y}$ for a counterflow diffusion flamelet with $a = 100 \, [1/s]$. 

FIGURE 6.6: Partially-premixed flamelets, in terms of $G$ and $\chi_{Z}$ at $Z = Z_{st}$, which are used to determine modeling coefficients $\alpha_1$ and $\alpha_2$ in switch function (6.34).

test data as a function of $G$ and $\chi_{Z}$ at the stoichiometric plane. Each point denotes a different flame configuration, in which open symbols correspond to steps in the applied strain rate and closed symbols correspond to steps in $\Phi_F$ and $\Phi_O$.

Data from partially premixed flamelets, henceforth indicated by PP, is used to compute normalized quantities

$$\theta = \left( \varphi^{PP} - \varphi^{NP} \right) / \left( \varphi^P - \varphi^{NP} \right) ,$$

in which $\varphi$ can denote $\dot{\omega}_{Y^*}, Y_i$ or $T$. Values are retrieved from the NP-FGM and P-FGM as a function of the $(Z^{PP}, Y^{PP})$ values from the PP flamelets. For all thermochemical variables which are retrieved from FGM databases, $\theta$ seems to obey a hyperbolic tangent-shaped transition from non-premixed ($\theta = 0$) to premixed ($\theta = 1$) combustion when displayed as a function of $\log_{10} \left( \chi_{Y^*} / \chi_{Z^*} \right)$ as can be seen in figure 6.7. The proposed switch function therefore is:

$$\Theta = \frac{1}{2} \left(1 + \tanh \left[ \alpha_1 \log_{10} \left( \chi_{Y^*} / \chi_{Z^*} \right) + \alpha_2 \right] \right) ,$$

in which $\alpha_i$ are modeling constants. For each individual variable, constants $\alpha_i$ are derived from partially-premixed flamelets by means of a least squares method. For the
6.4 Combination of FGM tables

computation of $\alpha_i$ only data for which $Y_i > 0.01Y_{i,\text{max}}$ and $\theta \in [-2, 3]$ have been taken into account; coefficients $\alpha_i$ are tabulated in table 6.2. A tabulated thermodynamic variable is now composed from the P-FGM and NP-FGM according to:

$$\varphi^{PP} = \Theta \varphi^P + (1 - \Theta) \varphi^{NP}.$$ (6.35)

Validation of proposed switch in limiting cases

The proposed switch function (6.34) reproduces the limiting premixed and non-premixed cases with only very minor errors as can be seen in figure 6.8. For the premixed flame with unit Lewis number $\chi_Z = 0$ everywhere. In premixed flames without differential diffusion effect $\chi_Z = 0$ implying that the switch (6.34) becomes undetermined; in computations $\Theta$ is set to 0.5 when $\chi_Z$ drops below a numerical threshold value ($\chi_Z < 10^{-6}$). Because thermochemical variables from the P-FGM and NP-FGM are identical in the unburnt mixture and nearly identical in the burnt state, the progress variable error $\epsilon_3$ in spatial coordinates, defined as
the combined FGM approach with optimized coefficients \( \alpha \). The vertical dashed line at \( Z = 0.2 \) in figures on the right indicates the scale change of the horizontal axis. The dash-dotted line denotes error (6.36).

\[
\epsilon_x (x) = \left| \frac{Y_{\text{FGM}}(x) - Y_{\text{DET}}(x)}{Y_{\text{DET}}(x)} \right| ,
\]

(6.36)

generally remains smaller than \( 10^{-3} \) for these premixed flames.

Due to numerical discretization and interpolation inaccuracies in generation of, and data retrieval from FGM tables, small non-zero values are obtained for \( \chi_y \), in a non-premixed flame. From the right graph of figure 6.5 it can be seen that the applied correction for the scalar dissipation rate for the progress variable, \( \epsilon \), the replacement of \( \chi_y \) by \( \chi_{y*} \), results in an error reduction of two to three orders of magnitude for a non-premixed flame. \( \chi_Z \) is of the same order of magnitude as \( \chi_y \), implying that for a non-premixed flame the ratio \( \chi_{y*}/\chi_Z \sim O(10^{-2}) - O(10^{-3}) \). Application of the switch function results in good predictions of flame structure, depicted in the right part of figure 6.8. For these non-premixed flames \( \epsilon_{y*} \), which is now defined as a function of \( Z \), remains smaller than \( 10^{-3} \) but tends to increase slightly with increasing applied strain rate.
It is concluded from figure 6.8 that the proposed switch function (6.34) recognizes the two limiting cases for partially-premixed flames, premixed and non-premixed flames, very well and errors in flame structure remain negligible. In the subsequent section, partially-premixed flames are computed using the proposed combined FGM approach and the accuracy of predictions is determined by comparison with flames computed using detailed chemistry.

### 6.5 Combined FGM approach in partially-premixed flames

The proposed switch function (6.34) is used to compute partially-premixed flames in a counterflow set-up. Different stratification levels are prescribed in terms of $\Delta \Phi$; boundary conditions are summarized in table 6.3. The least stratified counterflow flame ($\Delta \Phi = 0.6$) closely corresponds to flames for which has been shown [Fiorina2005-1, Bongers2005-2] that a P-FGM yields accurate predictions for the progress variable profile. The other stratification level presented in this study ($\Delta \Phi = 0.8$) extends well beyond the flammability limits for premixed flames.

#### Flame structure

In general, flame structures of the partially-premixed flames are quite well predicted by both the P-FGM and the NP-FGM having $Z = 0$ and $Z = 1$ as boundary conditions. From the right graph in figure 6.1 can be seen that in the P-FGM non-zero $\dot{\omega}_Y$-values are obtained closer to the stoichiometric plane than in the NP-FGM. This explains why the maximum $Y$ value in the partially-premixed flames, see figure 6.9, is generally found at lower $Z$-values when P-FGM results are compared to NP-FGM outcomes. Figure 6.9 shows as well that for small applied strain rates the P-FGM reproduces detailed chemistry results very well while the NP-FGM overestimates $Y$ in rich conditions. When the applied strain rate is increased, the P-FGM starts to deviate from detailed chemistry results by an underestimation of $\dot{\omega}_Y$. At the same time, the NP-FGM tends to become more accurate with increasing applied strain rate. At high applied strain rates the NP-FGM yields significantly better results than the P-FGM. For a fixed applied strain rate, an increased stratification leads to a slightly better performance of the NP-FGM and a minor deterioration of the P-FGM predictions.

The combined FGM approach using switch function (6.34) results in a $Y$-profile which lies in between the obtained profiles using either the P-FGM or NP-FGM. For intermediate applied strain rates, where the P-FGM underestimates $Y$-values while the NP-FGM overestimates them, the combined FGM approach obtains the best resemblance with detailed chemistry data: it outperforms both the P-FGM and NP-FGM approach. For either very low or very high applied strain rates the combined FGM obtains better results than the least applicable type of FGM, but results are not as good as obtained by using the

<table>
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<tr>
<th>$\Delta \Phi$</th>
<th>$Z_{ox}$ [-]</th>
<th>$Z_{fu}$ [-]</th>
<th>$\phi_{ox}$ [-]</th>
<th>$\phi_{fu}$ [-]</th>
<th>$a_{ext}$ [1/s]</th>
</tr>
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<tbody>
<tr>
<td>0.6</td>
<td>$1.44 \times 10^{-2}$</td>
<td>$1.89 \times 10^{-1}$</td>
<td>0.25</td>
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<tr>
<td>0.8</td>
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<td>$3.44 \times 10^{-1}$</td>
<td>0.11</td>
<td>9.00</td>
<td>982</td>
</tr>
</tbody>
</table>

**Table 6.3:** Boundary conditions and extinction strain rates for partially premixed one-dimensional counterflow flames used in this study.
Figure 6.9: Structure of progress variable in mixture fraction space in partially premixed flamelets with a low stratification of $\Delta \Phi = 0.6$ (left) and a high stratification of $\Delta \Phi = 0.8$ (right). Applied strain rates equal 20, 200 and 800 s$^{-1}$ (top to bottom). Circles indicate the detailed solution, solid lines indicate results obtained using a P-FGM, dashed lines indicate results obtained using a NP-FGM and solid lines with square markers indicate results obtained using the proposed combined FGM approach. The vertical dashed line indicates the stoichiometric $Z$-value.

Species mass fractions

As pointed out in section 6.1, accurate predictions for $\text{H}_2$ and CO are still hard to obtain for partially-premixed flames. In this section it is assessed whether the combined FGM approach can improve mass fraction predictions for these species when compared to either a P-FGM or NP-FGM. Results are presented in $Z$-space to exclude flame displacement effects and a priori results are added to estimate the influence of the error in $Y$ when an FGM table is used to represent combustion chemistry. For the a priori analysis values for $\chi_Z$ and $\chi_Y$ are reconstructed from the detailed chemistry solution and subsequently species mass fractions are retrieved making use of switch function (6.34). Whenever an FGM table is used in a computation, interpolation errors made during table generation and table usage can influence the computation outcome. However, figure 6.2 shows that $\varepsilon_Y$, as defined by equation (6.26), is smaller than $4 \times 10^{-4}$ for a non-premixed counterflow flame simulation in which a $401 \times 401$ ($Z \times Y$) FGM table with identical fuel and oxidizer
composition is used. Since the same FGM table resolution is used in the current one-dimensional flame simulations, it is concluded that interpolation errors are negligible to errors originating from inaccurate representation of partially-premixed combustion chemistry by FGM tables based on either premixed or counterflow diffusion flamelets.

Figure 6.10 shows that the P-FGM yields fairly accurate predictions but has the tendency to overestimate OH mass fractions for \( Z_{st} < Z < 1.5Z_{st} \). Predictions using the NP-FGM are even better than computation outcomes using the P-FGM. The proposed switch function performs very well: in both the lean and rich conditions detailed results are reproduced very well. For high applied strain rates \( a \) \textit{a priori} results approach detailed chemistry results better than any FGM approach, indicating that the error in OH mass fraction predictions can be attributed mainly to errors in \( Y \)-values which are visualized in figure 6.9. These results are analogous to those presented in section 4.3.1 and 4.3.2.

Figure 6.11 shows that \( \text{H}_2 \) mass fraction predictions from a P-FGM and NP-FGM differ significantly in rich conditions. For lean and stoichiometric conditions both FGM tables yield predictions which are very close to detailed chemistry results. In rich conditions, the P-FGM consistently overestimates the \( \text{H}_2 \) mass fraction which is explained by the fact that diffusion is only allowed in \( Y \)-direction in premixed flamelets. In non-premixed flamelets...
**Figure 6.11:** H$_2$ mass fraction in Z-space in partially premixed flamelets with a stratification of $\Delta \Phi = 0.6$ (left) and $\Delta \Phi = 0.8$ (right). Applied strain rates equal 20, 200 and 800 [1/s] (top to bottom). Symbols are identical to those in figure 6.10.

H$_2$ diffuses towards the stoichiometric plane where it is consumed resulting in lower H$_2$ mass fractions. The NP-FGM slightly underestimates H$_2$ values for $Z \geq 2Z_{st}$. The combined FGM approach does a much better job than the P-FGM and outperforms the NP-FGM for $Z \geq 3Z_{st}$. For flames in which the $Y$-profile using the combined FGM approach significantly deviates from detailed chemistry results, e.g. for $a = 20$ and $a = 800$ [1/s], *a priori* results tend to be worse than the combined FGM approach, indicating a (partial) cancelation of errors: in the combined FGM approach the error in $Y$ has a beneficial effect on H$_2$ mass fraction predictions. As depicted in the left graph of figure 6.13, in the P-FGM the H$_2$ mass fraction increases rapidly with increasing $Y$ close to chemical equilibrium, i.e. $C = 1$, under rich conditions. By replacing the $Y$-values obtained by combined FGM approach by the slightly higher values from the detailed chemistry computation, the H$_2$ mass fraction contribution from the P-FGM increases significantly.

For CO a similar behavior as for H$_2$ is observed with one major difference: *both* the P-FGM and the NP-FGM overestimate CO mass fractions for rich conditions, although in the NP-FGM results this overestimation is less severe. Because the combined FGM approach combines data from the P-FGM and the NP-FGM, results for this approach will always be better than the P-FGM results but never be as good as the NP-FGM simulation outcomes. *A priori* results for CO are slightly better at low applied strain rates, but worsened at high applied strain rates. At high strain rates the inverse occurs: $Y$-values from the detailed
Figure 6.12: CO mass fraction in Z-space in partially premixed flamelets with a stratification of \( \Delta \Phi = 0.6 \) (left) and \( \Delta \Phi = 0.8 \) (right). Applied strain rates equal 20, 200 and 800 [1/s] (top to bottom). Symbols are identical to those in figure 6.10.

chemistry simulations are higher than for the combined FGM approach. Like H\(_2\), the CO mass fraction increases rapidly with increasing \( Y \) close to chemical equilibrium under rich conditions. This causes the worsening of CO mass fraction predictions when \( Y \)-values from the combined FGM simulation are replaced by detailed chemistry values, as done in the \textit{a priori} analysis.

### 6.6 Transport equations for species with large timescales

In the previous section, it has been observed that CO and H\(_2\) are predicted far less satisfactory than the radical species OH. From figure 6.14 can be observed that for OH production and consumption rates are orders of magnitude larger than the net production rate, implying a small associated chemical timescale. For CO the production and consumption rates are an order of magnitude smaller than for OH and while its maximum mass fraction is an order of magnitude larger. This indicates that the chemical timescale of CO-formation is much smaller than the one of OH-formation. The less satisfactory predictions for CO might thus be caused by a trajectory in composition space of the partially-premixed flames that does not lie on low-dimensional manifolds constructed from either premixed or non-premixed flamelets. CO can be allowed to depart from the manifold by solving a transport equation for this species, instead of directly interpolating
its mass fraction from FGM tables. The one-dimensional transport equation for a species $i$ in a counterflow set-up reads:

$$\frac{\partial \rho Y_i}{\partial t} + \frac{\partial \rho u Y_i}{\partial x} = \frac{\partial}{\partial x} \left( \frac{\lambda}{\epsilon_p} \frac{\partial Y_i}{\partial x} \right) + \dot{\omega}_{Y_i} - \rho Y_i G,$$

in which the chemical source term $\dot{\omega}_{Y_i}$ is retrieved from the combined manifolds in an identical way as other thermochemical variables, i.e. by function (6.34). Solving an additional transport equation, and the inherent effect that $Y_i$ now can attain other values than would be obtained by direct interpolation from FGM tables, does not affect the manifold. Only the value for $Y_i$ is altered in this approach, which is a one-way coupling.

**Figure 6.13:** Hydrogen mass fraction $Y_{\text{H}_2}$ (left) carbon-monoxide mass fraction $Y_{\text{CO}}$ (right) from the P-FGM. $C$ denotes the normalized progress variable $C = Y/Y_{eq}$. Only the $Z$-range of significance has been shown; every second entry in $Z$-direction and every eighth entry in $C$-direction is shown.

**Figure 6.14:** Production and consumption rates of OH (top) and CO (bottom) in $Z$-space in partially premixed flames with a stratification of $\Delta \Phi = 0.6$ (left) and $\Delta \Phi = 0.8$ (right). In all flames the applied strain rate equals 200 [1/s]. Solid lines with solid markers indicate production rates, solid lines with open markers indicate consumption rates and solid lines without markers indicate net production rates. The thin vertical dashed line indicates the stoichiometric mixture fraction value. Only the part of the $Z$-space where non-zero values are obtained is shown.
Figure 6.15: Structure of CO mass fraction in mixture fraction space in partially premixed flamelets with a low stratification of $\Delta \Phi = 0.6$ (left) and a high stratification of $\Delta \Phi = 0.8$ (right). Strain rates equal 20, 200 and 800 s$^{-1}$ (top to bottom). Solid line with square markers denote direct (mass fraction) retrieval from FGM tables, solid line without makers denotes transport equation solution.

between the chemical manifold and the flame simulation just as well as the direct interpolation approach. Conservation of mass is thereby no longer guaranteed: constraint (1.30) may thus be violated by this approach.

Figure 6.15 shows that the approach of solving an additional transport equation, in which the chemical source term is a weighted combination of source terms from P-FGM and NP-FGM tables, can improve results, especially at intermediate strain rates and in rich conditions. For an applied strain rate of 200 [1/s], CO mass fractions are predicted much better for $Z > Z_{\text{st}}$ by solving an additional transport equation instead of direct interpolation. For the other applied strain rates, generally an improvement in predictions (or at least no significant deterioration) is observed for $Z > Z_{\text{st}}$ as well. In the burn-out zone, i.e. close to chemical equilibrium, the oxidation of CO to CO$_2$ on the lean side of the stoichiometric plane, yields a negative net CO formation rate in this region. This explains why (non-physical) negative mass fraction values are obtained at lean conditions for small values of the applied strain rate $a$. It is concluded that introduction of an additional transport equation is no general applicable method to improve CO mass fraction predictions; under certain conditions results improve while in other regions non-physical values are obtained. The regions where the most significant improvements have been
obtained, are diffusion-dominated regions with $Z > Z_{st}$.

6.7 Conclusions

Accurate predictions for $\text{H}_2$ and CO in partially-premixed flames are still hard to obtain, evidenced by publications devoted to this subject, both on laminar flames [Bongers2005-2, Fiorina2005-1] and turbulent flames [Bradley2002, Vervisch2004, Vreman2008]. Chapter 4 pointed out that a dedicated FGM table with the right boundary conditions can result in fairly accurate predictions, but for real-life applications such boundary conditions are not always available. In this chapter a novel approach was presented which combines premixed and non-premixed flamelet-based FGM tables based on local scalar dissipation rates. Validation of this approach has been performed by simulating the two limiting cases in partially-premixed combustion: a premixed and a non-premixed flame. Both flames were very accurately reproduced: maximum values for the error in $Y$, defined by equation (6.36), were three orders of magnitude smaller than absolute $Y$-values.

When the proposed switch function (6.34) is applied to one-dimensional partially-premixed flames, their flame structure is quite well reproduced by the combined FGM method. For low and high applied strain rates, the error is only slightly larger than the error obtained using only the FGM table (premixed flamelet-based or counterflow diffusion flamelet-based) which yields the most accurate predictions, while for intermediate cases the combined approach yields the best results.

OH mass fractions are very well predicted by the combined FGM approach: the overestimation of OH by the P-FGM for slightly rich conditions is not observed in combined FGM results. Outcomes of an a priori analysis indicate that the combined FGM approach reproduces OH mass fractions very well, for high applied strain rates the major cause for deviations is the underestimation of $Y$-values. $\text{H}_2$ mass fractions are well predicted by the combined FGM approach: the notorious overestimation of $\text{H}_2$ by the P-FGM is, to a great extend, balanced by the combination with the NP-FGM, which slightly underestimates $\text{H}_2$. At low applied strain rates and for $Z \gtrapprox 3Z_{st}$ the combined FGM approach reproduces $\text{H}_2$ mass fractions better than either the P-FGM or the NP-FGM. Results of the a priori analysis indicate that these fairly good results are partly caused by a cancelation of errors: the error in $Y$ has a beneficial effect on $\text{H}_2$ mass fraction predictions for both low and high applied strain rates.

For CO the combined FGM approach does not obtain better results than the use of a single FGM: since both the P-FGM and the NP-FGM overestimate CO mass fractions for rich conditions, combining these two FGM tables will in the most favorable case yield identical results as the most applicable FGM table. A high applied strain rates, a similar cancelation of errors occurs for CO as for $\text{H}_2$: the underestimation in $Y$ has a beneficial effect on CO mass fraction predictions. The large chemical time scale of CO-formation, might suggest that errors can also be caused by the fact that CO is only weakly attracted to the chemical manifold. The introduction of an additional transport equation for CO, which takes into account its transient behavior, does significantly improve CO in rich conditions, especially at intermediate applied strain rates. However, for small values of the applied strain rate (non-physical) negative mass fraction values are obtained at lean conditions. The introduction of an additional transport equation for CO is thus no generally applicable
method to improve its mass fraction predictions.

It can be concluded that the combined FGM approach is a safe method to use when the degree of premixing in not known \textit{a priori}. Although results are not always as accurate as the best-performing (single) FGM table, simulation outcomes showed better correspondence with detailed chemistry results than the worst-performing FGM table for all cases discussed in this chapter.
Combined FGM approach
Summarizing conclusions

In this thesis the Flamelet Generated Manifold (FGM) method has been applied to partially-premixed turbulent flames with increasing stratification levels. The FGM reduction method greatly reduces both the number of equations to be solved, and the stiffness of the resulting system of equations. On top of these advantages, chemical source terms can be evaluated much faster than using the Arrhenius expression (1.21) for each elementary reaction. Thereby Large Eddy Simulation (LES) and Direct Numerical Simulation (DNS) of industry-relevant flames is enabled, which would have been impossible without the use of FGM or a comparable reduction technique. FGM tables are based on flamelet structures, and therefore an a priori choice has to be made for the type of flamelet before the FGM table can be constructed and subsequently used in a flame simulation. For flames with a very moderate stratification, thereby remaining within flammability limits, previous research has pointed out that premixed flamelets are the obvious flamelet type to construct the FGM table from. For flames treated in chapter 3, the FGM table could thus without debate be generated from premixed flamelets. For flames with increasing stratification, the choice what type of flamelet to use, could not be answered that easily, as pointed out in chapter 4.

For all partially-premixed flames treated in this thesis, FGM tables are parameterized on mixture fraction and progress variable to capture both mixing and reaction progress. Solving the flamelet equations for different boundary/initial conditions enables the generation of the two-dimensional FGM tables. For increased accuracy, additional controlling variables can be used but this generally leads to very large memory requirements. It is important for other applications that, although only the GRI 3.0 reaction mechanism is used in this thesis, the FGM reduction method imposes no restriction on the use of more detailed reaction mechanisms or fuel/oxidizer composition. However, during the research described in this thesis, it turned out that FGM creating is never just “plug-and-play” but requires the research engineer to reflect on modeling assumptions for his specific application. Procedures and software to generate FGM tables have matured to a level that enables distribution, but modeling assumptions for the used flamelet structures determine the accuracy, that can be obtained using the eventual FGM tables, to a great extend.

In chapter 3 DNS of the turbulent stratified Bunsen flames was carried out with the use of the FGM reduction method. Three stratification setups were used in which the mixing length scale was altered by decreasing the stratification length scale or increasing the range of equivalence ratio $\phi$. The extension of the range of $\phi$, besides the reduced mixing length scale, also leads to a larger variation in the mass burning rate $m$. Since the reaction layer must be resolved using the FGM approach, a large number of grid points is needed. The purpose of this study, was to analyze the applicability of the Flame Surface Density (FSD)
approach, which can relieve numerical requirements by orders of magnitude because the reaction layer no longer has to be resolved. The envisioned application of the FSD model is LES of turbulent partially-premixed flames. The FSD model has been developed specifically for premixed flames, the novelty of this work is the analysis of the FSD model for flames with a spatial variation in $\phi$. All studied flames were in the Thin Reaction Zones regime [Borghi1988] and flame structures observed in the DNS are nearly identical to premixed flamelets. Mean mixing length scales were approximately one order of magnitude larger than the flamelet flame thickness for ($\phi = 0.7$).

The FSD model proved to be applicable to stratified flames when the variation in $m$ with $\phi$ is taken into account. For small filter widths, $\Delta F/\delta L = O(10^0)$, the FSD model showed some deviation from DNS results due to clustering of the entire chemical source in an infinitely thin flame sheet. For these filter widths, the subfilter variation of $m$ can be adequately modeled using a Dirac-PDF for the subfilter $Z$-distribution. The discrepancies between the FSD model and DNS results become less prevalent when larger filter widths are considered. For $\Delta F/\delta L = O(10^1)$ good agreement between the model and DNS results was obtained. However, due to a larger subfilter $Z$-variation, the replacement of the Dirac-PDF by a $\beta$-PDF significantly improves predictions for the surface-averaged value of $m$.

The $a$ priori analysis implied that the flame surface density $\Sigma$ was extracted from the DNS results. When $\Sigma$ is modeled by a simple algebraic Bray-Moss-Libby (BML) type model, source terms for $0.9 \leq \tilde{C} \leq 1.0$ are slightly overestimated while high source term values are underestimated due to the limited maximum $\Sigma$-value dictated by the BML type model. It can be concluded that the FSD-model, using a $\beta$-PDF to account for subfilter fluctuations in $Z$, the modeled surface-averaged mass burning rate is in good agreement with the filtered source term obtained from DNS data. Even when the extracted flame surface is replaced by a very simple algebraic model for $\Sigma$, a fairly good agreement between the modeled and extracted chemical source is obtained. The implementation of the FSD model using mass burning rate data from flamelets can therefore be considered to be a feasible possibility for the simulation of turbulent stratified flames in an industrial environment. For future research it is recommended that a FSD model is implemented in an LES code and an assessment is made whether the most substantial error originates from the model for $\Sigma$ or the $\beta$-PDF model for the surface-averaged value of $m$. This assessment would point out which submodel is the bottleneck in simulations of partially-premixed flames.

The stratified Bunsen flames in chapter 3 have a relatively small variation in $\phi$. Chapter 4 shifts towards well-known jet flames having a significantly larger range in $\phi$: the Sandia Flames C to F. For these partially-premixed flames, the choice which flamelets to use for the generation of FGM tables is no longer an obvious one: although most publications point out that counterflow diffusion flamelets most closely correspond to experimental results, other authors [Bradley2002] hypothesize that especially for Flame F the use of premixed flamelets might result in improvements in predictions. To exclude numerical and modeling errors originating from Computational Fluid Dynamics (CFD) solvers, an $a$ priori analysis is carried out to asses the accuracy of species mass fractions predictions by the FGM table. Counterflow diffusion flamelet-based tables proved to be significantly more accurate than premixed-based ones for $H_2$, $CO_2$ and $CO$ mass fraction predictions, regardless of the used molecular transport model. For fuel-rich conditions premixed flamelet-based FGM tables tend to severely overestimate $H_2$ and $CO$ mass fractions while underestimating $CO_2$ mass fractions. Differential diffusion effects are only visible in regions with small turbulence.
intensities: for the measurements of the Sandia Flames, flamelet solutions with unit Lewis numbers for all species yield a better correspondence with measured quantities than flamelets with a complex molecular transport model.

A counterflow diffusion flamelet-based table, in which unit Lewis numbers have been assumed for all species, can accurately predict $\text{H}_2\text{O}$, $\text{H}_2$, $\text{CO}_2$ and $\text{CO}$ mass fractions for the Sandia Flames C to F. For OH this FGM table yields a good correspondence between averages of large ensembles of measured and predicted values but for individual measurements the standard deviation is large due to measurement uncertainties (10%) and reconstruction uncertainties for $Z$ and $\mathcal{Y}$. The accuracy that can be reached with a two-dimensional ($Z$-$\mathcal{Y}$) steady counterflow diffusion flamelet-based table using unit Lewis numbers is striking. Even for Sandia Flame F, which is known to exhibit significant extinction and re-ignition events, the predictions are in close agreement with experimental observations.

In CFD simulations of flames, additional errors to those originating from combustion chemistry representation by an FGM table occur. Chapter 4 has shown that an FGM table based on counterflow diffusion flamelets in which unit Lewis numbers are assumed for all species, obtains best correspondence with measurements in an a priori analysis. Even for notorious species like CO and $\text{H}_2$, accurate predictions were obtained. Computations of CO and $\text{H}_2$ mass fractions in Large Eddy Simulations (LES) of the Sandia flames D and F have been compared with measurements. The flow field, and as a consequence the field of passive scalar $Z$, are not very well recovered in the current simulations. This is attributed to the prescribed turbulent inflow field, which is a poor representation of the inflow conditions in the experiments. However, when $\mathcal{Y}$, $Y_{\text{CO}}$ and $Y_{\text{H}_2}$ results from simulations are depicted as a function of $Z$ instead of spatial coordinate, it is observed that for Flame D the flame structure is fairly well recovered. For Flame F, which exhibits significantly more non-equilibrium effects, the flame structure is less satisfactory recovered due to an overestimation of the progress variable source term in rich conditions. For both flames the inclusion of a Presumed PDF subfilter combustion model, using a $\beta$-PDF in both $Z$- and $\mathcal{Y}$-direction, does not significantly influence numerical result. Predictions for $Y_{\text{CO}}$ and $Y_{\text{H}_2}$ slightly improve in rich conditions when a transport equation, using the species its source term from the FGM table, is solved instead of directly interpolation from the FGM table. However, this approach leads to negative mass fraction predictions for both CO and $\text{H}_2$ in lean conditions. When errors in control variables $Z$ and $\mathcal{Y}$ are excluded by means of an a priori analysis, is found that results match experimental data much better than LES results. The relatively smooth behavior of $\text{H}_2$ in the FGM table makes accounting for variances of $Z$ and $\mathcal{Y}$ in the a priori analysis superfluous; for accurate a priori results for CO these variances should be taken into account. The discrepancy between a priori outcomes and LES results points out that a better description of boundary conditions for turbulence in the simulations probably results in better LES results, thereby making other expensive methods like the Lagrangian Flamelet Model [Pitsch2000] superfluous.

In chapter 6 a novel approach was presented which combines premixed and non-premixed flamelet-based FGM tables based on local scalar dissipation rates. The proposed method accurately reproduced a premixed and non-premixed flamelet solution with an error in $\mathcal{Y}$ three orders of magnitude smaller than the maximum $\mathcal{Y}$-value. The proposed combined FGM method quite well reproduced one-dimensional partially-premixed flame structures. For low and high applied strain rates, the error is only slightly larger than the error
obtained using the (single) FGM table which reproduces best the flame structure; it has to be stressed that this is an a posteriori comparison here. For intermediate applied strain rates the combined approach yields the best results. H\textsubscript{2} mass fractions are well predicted by the combined FGM approach: the notorious overestimation of H\textsubscript{2} by the FGM-P is, to a great extend, balanced by the combination with the FGM-NP, which slightly underestimates H\textsubscript{2}. At low applied strain rates and for Z \gtrsim 3Z\textsubscript{st} the combined FGM approach reproduces H\textsubscript{2} mass fractions better than either the FGM-P or the FGM-NP. Results of the a priori analysis indicate that these fairly good results are partly caused by a cancelation of errors: the error in \(Y\) has a beneficial effect on H\textsubscript{2} mass fraction predictions for both low and high applied strain rates. For CO the combined FGM approach does not obtain better results than the use of a single FGM: since both the FGM-P and the FGM-NP overestimate CO mass fractions for rich conditions, combining these two FGM tables will in the most favorable case yield identical results as the most applicable FGM table. At high applied strain rates, a similar cancelation of errors occurs for CO as for H\textsubscript{2}: the underestimation in \(Y\) has a beneficial effect on CO mass fraction predictions. Without this underestimation of \(Y\)-values, predictions for CO would worsen by the contribution of the FGM-P in which CO increases rapidly with increasing \(Y\) for rich conditions. The introduction of an additional transport equation for CO, which takes into account its transient behavior, does significantly improve CO in rich conditions, especially at intermediate applied strain rates. However, for small values of the applied strain rate, negative mass fraction values are obtained at lean conditions. As was also pointed out in chapter 5, the introduction of an additional transport equation is no generally applicable method to improve species mass fraction predictions.

Finally, it should be noted that the FGM method enables DNS and LES of complex flames which, without this or a comparable reduction method, would have taken unavailable amounts of computational resources. The FGM approach requires the user to have at least a basic understanding of flamelets and is not straightforward to implement for applications in which the range in equivalence ratio present in the combustor is unknown. Nevertheless, the author believes that a working knowledge of flamelets is not hard to obtain and a basic insight in combustion processes should suffice and enable the FGM user to define the appropriate flamelet type and its boundary conditions for the simulation of partially-premixed flames. The combined FGM approach as proposed in chapter 6 needs to be developed further, but would reduce the required user input thereby making the method more robust. For applications in which auto-ignition occurs it might be useful to include a table based on homogenous reactors in the combined FGM approach. This table should be used instead of flamelet-based tables when both the gradient in \(Z\) and \(Y\) is small, indicating that diffusive processes do not play an important role and no typical flamelet structure can be identified in the multi-dimensional flame.
Nomenclature

Latin

$A_j$ Pre-exponential factor
$a$ Applied strain rate
$C$ Normalized progress variable
$c_{v,i}$ Specific heat of species \( i \) at constant volume
$c_v$ Weighted specific heat at constant volume
$c_{p,i}$ Specific heat of species \( i \) at constant pressure
$c_p$ Weighted specific heat at constant pressure
$Da$ Damkohler number
$D_i$ Mixture-averaged diffusion coefficient
$D_{ij}$ Binary diffusion coefficient
$E_A$ Activation energy
$G$ Flame stretch rate
$g$ Gravitational acceleration vector
$h$ Enthalpy
$h_{i}$ Enthalpy of species \( i \)
$I$ Unit tensor
$K$ Mass based flame stretch rate
$k_{eq}^j$ Equilibrium constant of reaction \( j \)
$k_f^j$ Forward reaction rate of reaction \( j \)
$k_r^j$ Reverse reaction rate of reaction \( j \)
$L_{INT}$ Integral length scale
$L_{MIX}$ Mixing length scale
$Le_i$ Lewis number of species \( i \)
$m$ Mass burning rate
$M$ Mean molecular mass
$M_i$ Molecular mass of species \( i \)
$m$ Mass burning rate
$N_e$ Number of elements
$N_r$ Number of reactions
$N_s$ Number of species
$N_G$ Number of grid points
$N_T$ Number of time increments
$n$ Normal vector
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* The units of the reaction rate variables depend on the reaction order. The unit of the pre-exponential factor depends on the reaction constant $\beta$ as well.

**Greek**

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<tbody>
<tr>
<td>$\phi$</td>
<td>Variable stored in FGM database</td>
</tr>
<tr>
<td>$\chi$</td>
<td>Scalar dissipation rate</td>
</tr>
<tr>
<td>$\omega_i$</td>
<td>Nett chemical source of species $i$</td>
</tr>
<tr>
<td>$\omega_i^+$</td>
<td>Total chemical production of species $i$</td>
</tr>
<tr>
<td>$\omega_i^-$</td>
<td>Total chemical consumption of species $i$</td>
</tr>
</tbody>
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Sub- and superscripts

<table>
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<tr>
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<tr>
<td>$-\infty$</td>
<td>Initial</td>
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<td>$b$</td>
<td>Burnt</td>
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<td>$\text{eq}$</td>
<td>Equilibrium</td>
</tr>
<tr>
<td>$f$</td>
<td>Forward</td>
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<td>$\text{fu}$</td>
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<td>$\text{st}$</td>
<td>Stoichiometric</td>
</tr>
<tr>
<td>$u$</td>
<td>Unburnt</td>
</tr>
</tbody>
</table>
Flame stretch transport equation

In section 2.2.2 a transport equation for local flame stretch rate $G$ has been introduced. In this appendix equation (2.38) is derived, which differs slightly from previous work [Oijen2002-2] because it contains a derivative with respect to time.

Since a counterflow flamelet is assumed to be flat and a planar geometry is considered only in this thesis, combustion variables $\rho, T, Y_i, \mu$ and $G$ only depend on the $x$-coordinate, perpendicular to the flame surface, and time $t$. The $x$-component of the gas velocity $u$ is a function of $x$-coordinate and $t$ but the $y$-component of the gas velocity $v$ and pressure $p$ are functions of both coordinates $x$ and $y$ and $t$. When the definition for $G$, i.e.

\[ G(x, t) = \frac{\partial v}{\partial y}, \]  

(A.1)

and continuity equation (2.37) are substituted in the two-dimensional momentum equation (1.27) in $x$-direction, one obtains [DixonLewis1990, Stahl1991]:

\[ \rho \frac{\partial u}{\partial t} + \rho u \frac{\partial u}{\partial x} + \frac{\partial p}{\partial x} = \frac{\partial}{\partial x} \left[ \frac{4}{3} \mu \frac{\partial u}{\partial x} - \frac{2}{3} \mu G \right] + \mu \frac{\partial G}{\partial x}. \]  

(A.2)

In a similar way, the two-dimensional momentum equation (1.27) in $y$-direction can be reduced to:

\[ \rho \frac{\partial v}{\partial t} + \rho u \frac{\partial v}{\partial x} + \rho v G + \frac{\partial p}{\partial y} = \frac{\partial}{\partial x} \left[ \mu \frac{\partial v}{\partial x} \right] + \frac{\partial}{\partial y} \left[ \frac{4}{3} \mu G - \frac{2}{3} \mu \frac{\partial u}{\partial x} \right]. \]  

(A.3)

When the definition for $G$, equation (A.1) is integrated with respect to $y$ the following expression is obtained:

\[ v(x, y, t) = y G(x, t) + A_1, \]  

(A.4)

in which the integration constant $A_1$ must equal zero because the velocity at the flamelet its centerline ($y = 0$) has no vertical component. Substitution of equation (A.4) in equation (A.3) yields the following expressions for the momentum equations in $x$- and $y$-direction:

\[ \frac{\partial p}{\partial x} = -\rho \frac{\partial u}{\partial t} - \rho u \frac{\partial u}{\partial x} + \frac{\partial}{\partial x} \left( \frac{4}{3} \mu \frac{\partial u}{\partial x} - \frac{2}{3} \mu G \right) + \mu \frac{\partial G}{\partial x}, \]  

(A.5)

\[ \frac{1}{y} \frac{\partial p}{\partial y} = -\rho \frac{\partial G}{\partial t} - \rho u \frac{\partial G}{\partial x} - \rho G^2 + \frac{\partial}{\partial x} \left( \mu \frac{\partial G}{\partial x} \right). \]  

(A.6)

From equation (A.6) can be seen that its left hand side can only be a function of $x$ and $t$ since it right hand side is. By taking the derivative of equation (A.5) with respect to $y$, and subsequent division by $y$, the following expression is obtained:
\[ \frac{1}{y} \frac{\partial}{\partial y} \left( \frac{\partial p}{\partial y} \right) = \frac{1}{y} \frac{\partial}{\partial x} \left( \frac{\partial p}{\partial y} \right) = \frac{\partial}{\partial x} \left( \frac{1}{y} \frac{\partial p}{\partial y} \right) = 0, \]  

(A.7)

implying that the left-hand side of equation (A.6) can merely be a constant or a function of \( t \) throughout the entire flow field. Subsequently, this term will be denoted by \(-P(t)\), which yields the final transport equation for \( G \):

\[ \rho \frac{\partial G}{\partial t} + \rho u \frac{\partial G}{\partial x} + \rho G^2 = \frac{\partial}{\partial x} \left( \mu \frac{\partial G}{\partial x} \right) + P(t). \]  

(A.8)

In a (counterflow) flamelet, \( P(t) \) is governed by the flamelet its boundary condition, as is further shown in section 2.2.2.
Bibliography


Bibliography


Curriculum Vitae

10/2011–currently  CFD Research Engineer  
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                  Obtained awards: KVGN Gasunie Thesis Prize and the IGU Thesis Prize

Publications following from this thesis


3. L.M. Verhoeven, W.J.S. Ramaekers, J.A. van Oijen and L.P.H. de Goey, Modeling non-premixed laminar co-flow flames using Flamelet Generated Manifolds, accepted by *Combust. Flame*
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