MODELING PREFERENTIAL DIFFUSION EFFECTS IN PREMIXED METHANE-HYDROGEN-AIR FLAMES BY USING FLAMELET-GENERATED MANIFOLDS

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Abstract. In this paper, we investigate the extension of the flamelet-generated manifold (FGM) method to include preferential diffusion effects in methane-hydrogen-air flames. The FGM technique is a method to simplify the chemical kinetics model, which has been proven accurate for the modeling many type of flames. Due to the high diffusivity of hydrogen, flames in methane-hydrogen blends have a characteristic cellular structure, which is caused by thermal-diffusive instabilities. To account for preferential diffusion effects an additional degree of freedom is used to describe the combined fluctuations in enthalpy and element mass fractions. This approach is used to perform direct numerical simulations of lean premixed flame kernels in a turbulent flow field. The results are compared with results of simulations employing the full chemistry model. Extension of the manifold with one dimension appears to be enough for accurate predictions of species concentrations and burning velocities.
1 INTRODUCTION

There is a growing interest in using hydrogen containing fuel blends for practical combustion applications. Combustion of hydrogen enriched fuel mixtures allows for leaner operation of premixed turbulent combustion and leads to reduced emissions and to higher efficiencies of engines or turbines. Due to the high diffusivity of hydrogen, flames in these mixtures have a characteristic cellular structure, which is caused by thermal-diffusive instabilities [1]. The modeling of preferential diffusion (or non-unity Lewis number) effects that cause these instabilities, is investigated in this paper in the context of direct numerical simulations (DNS) employing the flamelet-generated manifold (FGM) technique [2, 3]. The FGM technique is a method to simplify the chemical kinetics model taking the local flamelet structure into account. It has already proven to be accurate for modeling (partially) premixed flames in DNS, LES and RANS settings [3, 4]. Previous research has focused on flames with unity Lewis number transport models, thereby neglecting preferential diffusion effects. The FGM method is extended in the present contribution by introducing an additional mixing parameter in the manifold, describing the combined fluctuations in enthalpy and element mass fractions due to flame stretch and preferential diffusion.

The outline of this paper is as follows. In the next section, the FGM method is explained. Extra attention is given to the extension of the method to account for preferential diffusion effects. In the following section, the method is applied to laminar counterflow flames and expanding flame kernels in a turbulent flow field for validation. After that, the results are analyzed with respect to flame stretch and its influence on the burning velocity. The paper ends with conclusions.

2 FLAMELET-GENERATED MANIFOLDS

2.1 Flamelet description for premixed flames

In this section, the flamelet description for premixed flames introduced by De Goey and Ten Thije Boonkkamp [5], is briefly reviewed. It is has been derived in a systematic way by decomposing the system of combustion equations in three parts: 1) a flow and mixing part without chemistry, 2) a kinematic equation for the flame motion, including internal flame dynamics, and 3) a flamelet part describing the inner structure and propagation speed of the flame structure. The flame, including internal structure, is described in terms of iso-surfaces of a progress variable $\mathcal{Y}$, which can be any linear combination of species mass fractions. The motion of each iso-surface of $\mathcal{Y}$ is described by the kinematic equation [6]

$$\frac{\partial \mathcal{Y}}{\partial t} + \mathbf{v}_t \cdot \nabla \mathcal{Y} = \frac{\partial \mathcal{Y}}{\partial t} + \mathbf{v} \cdot \nabla \mathcal{Y} - s_L |\nabla \mathcal{Y}| = 0$$

(1)

where $\mathbf{v}_t = \mathbf{v} + s_L \mathbf{n}$ is the local velocity of a flame surface being the sum of the fluid velocity $\mathbf{v}$ and the local burning velocity $s_L \mathbf{n}$ and where the unit normal vector $\mathbf{n}$ on the flame surface, directed towards the unburnt mixture, can be written as $\mathbf{n} = -\nabla \mathcal{Y}/|\nabla \mathcal{Y}|$. 

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Note that (1) is introduced for each iso-surface in the complete 'flame' region where $0 < \mathcal{Y} < 1$. As a result $\mathbf{v}_f$ and $s_L$ are field variables, which vary throughout the flame region. In some other publications (e.g. [7]) $s_L$ is referred to as displacement speed.

The stretch field $K$ inside the flame zone is defined as the relative rate of change of the mass $M$ in a small part of the flame, enclosed by a small 'flame' volume $V$, moving with velocity $\mathbf{v}_f$:

$$K = \frac{1}{M} \frac{dM}{dt} \quad \text{with} \quad M = \int_{V(t)} \rho dV.$$  \hfill (2)

From this definition of $K$ and the kinematic equation (1) the following set of flamelet equations for the conservation equations of mass and $\mathcal{Y}$ can be derived rigorously in a orthogonal flame-adapted coordinate system [5]:

$$\frac{\partial}{\partial s} (\sigma m) = -\sigma \rho K,$$  \hfill (3)

$$\frac{\partial}{\partial s} \left( \sigma m \mathcal{Y} - \sigma \rho D_i \frac{\partial \mathcal{Y}}{\partial s} \right) = \sigma \dot{\omega}_i - \sigma \rho K \mathcal{Y},$$  \hfill (4)

where $s$ is the arc length through a flamelet, locally perpendicular to the iso-planes of $\mathcal{Y}$ and $\sigma$ the surface area through which the transport processes in the flame-adapted system take place. Note that the local curvature $\kappa$ of the flame surfaces is related to the surface area $\sigma$ by

$$\kappa = \nabla \cdot \mathbf{n} = -\frac{1}{\sigma} \frac{\partial \sigma}{\partial s}.$$  \hfill (5)

In (3) the mass burning rate $m = \rho s_L$ is introduced with $\rho$ the density of the gas mixture. $D_i$ and $\dot{\omega}_i$ are the diffusion coefficient and chemical source term of species $i$, respectively. The diffusive fluxes are assumed to be proportional to the gradient of the species mass fraction. The equations for the other species mass fractions $Y_i$ have a similar form as (4)

$$\frac{1}{\sigma} \frac{\partial}{\partial s} \left( \sigma m Y_i - \sigma \rho D_i \frac{\partial Y_i}{\partial s} \right) - \dot{\omega}_i = -\rho K Y_i + Q_i,$$  \hfill (6)

but with an extra term $Q_i$ defined as

$$Q_i = \frac{d Y_i}{d \tau} + \nabla \cdot (\rho D_i \nabla \| Y_i),$$  \hfill (7)

with $\nabla \|$ the gradient operator in direction parallel to the flame surface only. The first term on the right-hand side accounts for unsteady effects in the flame-adapted reference frame: $\frac{\partial}{\partial \tau} = \frac{\partial}{\partial t} + \mathbf{v}_f \cdot \nabla$. The second term accounts for diffusive transport processes due to the fact that iso-planes of different species are not parallel. These extra terms are small and neglected inside the laminar flamelet regimes [5]. The set of flamelet Eqs. (3), (4) and (6) together with a similar equation for the enthalpy, describes the internal flame structure, dynamics and the eigenvalue for the mass burning rate $m(s)$ for a flamelet with a particular $Q_i(s)$, stretch field $K(s)$, and curvature field $\kappa(s)$. 

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2.2 Flamelet-generated manifolds

The flamelet equations derived in the previous section are used to generate FGM’s. If we assume that all perturbations from local 1D flat flame behavior can be neglected, then we can use \( K = 0, \kappa = 0 \) (or equally \( \sigma = 1 \)), and \( Q_i = 0 \). The set of equations then describes an unstretched, 1D, flat flamelet: the most elementary premixed flame structure. Its solution \( Y_i(s) \) can be considered as a 1D manifold in composition space. This 1D FGM can be parameterized by a single controlling variable \( c_1 \). In [2] it is shown that the 1D FGM corresponds to the 1D ILDM at high temperatures, where chemistry is dominant. At moderate temperatures, molecular transport is important as well and the FGM is closer to compositions found in a premixed flame.

In turbulent flames large deviations from the 1D manifold may arise due to flame stretch and curvature effects. To take this into account the dimension of the manifold can be increased. A larger degree of freedom makes a better description of the composition possible and therefore it increases the accuracy of the model. A multi-dimensional FGM is generated by computing a series of flamelets, which form together a multi-dimensional surface in composition space. A multi-dimensional FGM can be generated by including terms in the flamelet equations, which have been neglected in the 1D FGM approach. In this way more physics is taken into account, which results in a more accurate description of the composition.

2.3 Flame stretch and preferential diffusion

In this study we focus on the combined effect of preferential diffusion and flame stretch. It is well known that this combined effect results in a local redistribution of the element mass fractions \( Z_j \) and enthalpy \( h \) (see, e.g., [5]). This may cause fuel-enriched zones in a lean flame, resulting locally in more intense burning. Depending on the effective Lewis number of the reactants this may result in thermal-diffusive flame front instabilities [1].

Since these stretch effects are not present in a 1D unstretched flame, their influence on chemistry is not taken into account in a 1D FGM. To account for these effects, the variations in element mass fractions and enthalpy should be included in the manifold. This can be done by solving the flamelet equations for varying \( Z_j \) and \( h \) of the reactants as is described in [8]. If one would include these variations independently, this would mean that \( N_e \) additional dimensions have to be added to the manifold, with \( N_e \) the number of elements.

For weak stretch we have shown [9] that the changes in element mass fractions and enthalpy are linearly coupled. As a result the variations can be represented by a single controlling variable implying that only one additional dimension is needed for the manifold. This is very beneficial from a numerical point of view, but it needs to be investigated whether such correlations between the element mass fractions and enthalpy also exist for more general cases.

To add a dimension to the manifold that accounts for preferential diffusion effects, flame
stretch can be modeled, for instance, with a constant stretch rate \( K = K_u \) in the flamelet equations (3, 4, 6). By applying a range of stretch rates \( \rho K_u \delta f / m^0 = \ldots, -0.1, 0, 0.1, \ldots \) a 2D manifold is created.

A different way to model flame stretch in the flamelet equations, is by assuming a constant curvature \( \kappa = \kappa_u \) (or equivalently \( \sigma = \exp(-\kappa_u s) \)) and a stretch rate related to that, i.e. \( K = \kappa_u m^0 / \rho \) with \( m^0 \) the mass burning rate of an unstretched flat flame. By computing a series of flamelets with different curvatures \( \kappa_u \delta f = \ldots, -0.1, 0, 0.1, \ldots \), a 2D manifold is generated.

These 2D manifolds have to be parameterized by a set of two controlling variables, \( c_1 \) and \( c_2 \). Any combination of species’ mass fractions that results in a smooth mapping \( Y_i = f(c_1, c_2) \), is a suitable choice. For the 2D manifolds used in this study, appropriate controlling variables are

\[
\begin{align*}
  c_1 &= \phi_{\text{H}_2\text{O}} + \phi_{\text{CO}_2} + \alpha \phi_{\text{H}_2} + (1-\alpha) \phi_{\text{CH}_4} \\
  c_2 &= Z_C + Z_H
\end{align*}
\]

with \( \phi_i = Y_i / M_i \), \( M_i \) being the molar mass of species \( i \), and \( \alpha \) the volume fraction of \( \text{H}_2 \) in the fuel.

3 VALIDATION OF FGM

3.1 Laminar counterflow flames

To validate the FGM method, a series of premixed counterflow flames in the so-called back-to-back configuration has been computed with different FGM tables and with detailed chemistry. Two fuel mixtures are considered: pure methane and a mixture of 60% methane and 40% hydrogen by volume. These fuels are burnt with excess air at standard conditions with an equivalence ratio, \( \phi \), of 0.7. The simulations have been performed with the code CHEM1D [10]. An overview of the results is given in fig. 1, in which the mass burning rate \( m \) of each flame is shown as function of the applied strain rate \( a \). The mass burning rate is evaluated at the inner layer, which is here defined as the position of maximum chemical reaction rate of fuel conversion.

With increasing strain rate the mass burning rate decreases for the pure methane case, but for the hydrogen blend it remains nearly constant. This behavior is caused by strong preferential diffusion effects in the hydrogen case, and it has been observed by many others. The 1D FGM cannot reproduce this effect, because it does not allow variations in the element composition and enthalpy. The 2D FGM’s give much more accurate results. The exact way how flame stretch has been modeled in the flamelet equations does not appear to be important for the resulting manifolds, since 2D FGM A and B give similar results. For the pure methane case, the improvement of adding a dimension to the manifold is less dramatic as for the hydrogen blend, because the effective Lewis number is close to one for methane-air mixtures.
Figure 1: Mass burning rate $m/m^0$ at the inner layer of premixed counterflow flames as function of the applied strain rate $\alpha$. Simulations are performed with detailed chemistry (symbols), 1D FGM (dashed), and 2D FGM’s based on constant stretch fields (A, solid) and curvature-derived stretch fields (B, dash-dotted) for pure methane (blue) and a 60%–40% methane-hydrogen fuel mixture (red).

### 3.2 DNS of expanding flame kernels in 2D vortical flow

The second test case described here, focuses on the interaction between vortical structures and the flame front as it occurs in turbulent flames. The different manifolds are applied to DNS of premixed expanding flame kernels. The results are compared with simulations employing detailed chemistry. In order to keep the computational cost of the detailed simulations affordable, the simulations are limited to two spatial dimensions.

Freely expanding flame kernels are modeled in a homogenous isotropic turbulent flow field. The fuel mixture of 60% methane and 40% hydrogen by volume is premixed with air at an equivalence ratio of $\phi = 0.7$. The initial velocity fluctuations equal $u'/s^0_L = 4$ with $s^0_L$ the burning velocity of a flat unstretched flame with respect to the unburnt mixture. The pressure is atmospheric and the temperature of the reactants is 300 K. The integral length scale equals $\ell_T/\delta_f = 2$ with $\delta_f = 0.5$ mm the laminar flame thickness. The turbulent Reynolds number is $Re = u'\ell_T/s^0_L\delta_f = 8$. For these settings, combustion takes place in the thin reaction zones regime. Details about the code can be found elsewhere [3, 11, 12].

In fig. 2 the evolution of the flame computed with detailed chemistry is shown in terms of the mass fraction of the hydrogen radical $Y_H$. The initially spherical flame is perturbed by the vortices and finger-like structures arise pointing into the reactants. In these positively stretched regions of the flame, $Y_H$ is significantly higher than in the
Figure 2: Snapshots of the mass fraction of hydrogen radicals in a two-dimensional DNS of a spherically expanding methane-hydrogen flame kernel computed with detailed chemistry. Spatial dimensions are in millimeters. The color range indicates values from 0 to $1.4 \times 10^{-4}$. 
unperturbed initial condition. These regions correspond to locally richer zones, which burn more intensively. The regions in between the fingers are characterized by the opposite effect: lower $Y_H$ and fuel consumption rates (not shown).

The same simulations are performed with a 1D FGM and a 2D FGM based on stretched flamelets with constant curvature (FGM B). The results are compared in fig. 3. The distribution of both controlling variables and $Y_H$ are shown at $t = 0.36$ ms. It can be seen that the 2D FGM yields more accurate results than the 1D FGM. Since all combustion variables in the 1D FGM are solely determined by $c_1$ no variations of $c_2$ or $Y_H$ can be observed along the flame front. The additional degree of freedom in the 2D manifold allows these variations and the 2D FGM predicts very accurately the concentration of the hydrogen radical, which plays a key role in the chemistry of hydrocarbons.

In fig. 4 scatter plots are shown for the same fields as shown in fig. 3. Large fluctuations can be observed for $H_2$. At $c_1 = 0.85$, which is close to the inner layer, the difference between the maximum and minimum value of $Y_{H_2}$ is a factor of two. This implies a significant change in local stoichiometry, resulting in similar changes in mass burning rate. The variations in $Y_{H_2}$ are strongly correlated with the variations in the second controlling variable. Moreover, the combinations correspond to the 2D FGM, which indicates that a 2D manifold suffices.

4 FLAME STRETCH ANALYSIS

The combined effect of flame stretch and preferential diffusion result in local changes in element mass fractions and enthalpy. These changes on their turn result in changes in the local mass burning rate. To investigate this a similar analysis has been performed as in our previous work [3]. In this analysis 1D flame paths are reconstructed from the DNS results. Along these flame paths, flame stretch, curvature, and mass burning rate can be computed. By integrating the stretch profiles an integral Karlovitz number, $K_a$ can be obtained. In fig. 5, the mass burning rate at the inner layer of 1200 flamelets in the DNS result at $t = 0.6$ ms, has been plotted as function of the integral Karlovitz number. For unity Lewis number flames, this would result in scatter points on a line with slope of minus one. However, due to preferential diffusion effects deviations will arise. It can be seen that the scatter cloud has a smaller slope, corresponding to an effective Lewis number smaller than one. This is in agreement with theoretical models. However, the effect is not as large as for simulations of 1D flames with a stretch rate that corresponds to constant curvature. Moreover, if we compare these results with the slope of the results in fig. 1 for the counterflow flames, it can be seen that for the counterflow flames the slope is nearly zero. This difference is caused by the different stretch profiles. Note that the Karlovitz number is an integral of the stretch rate. Therefore, different stretch profiles may lead to the same Karlovitz number, but to different changes in $Z_j$ and $h$. This can be observed in the right plot of fig. 5, where these changes are plotted versus Karlovitz number. Very recently this was also observed in an analysis of results of a DNS of statistically flat flames [13].
Figure 3: Contour plots of progress variable (top row), second controlling variable (middle row), and mass fraction of hydrogen radicals (bottom row) computed with detailed chemistry (left), 1D FGM (middle), and 2D FGM B (right) at $t = 0.36$ ms. Spatial dimensions are in millimeters.
Figure 4: Scatter plot of hydrogen mass fraction versus scaled progress variable $c_1$ (left) and a scatter plot of hydrogen mass fraction versus second controlling variable conditioned at $c_1 = 0.85$. The solid blue circle and curve correspond to the 1D and 2D FGM, respectively.

Figure 5: Mass burning rate $m/m^0$ (left) and changes in element fractions $\Delta \psi_j = \Delta Z_j / M_j$ (right) at the inner layer against Karlovitz number for flamelet structures in the turbulent flame kernel (symbols) and for flamelets with stretch rates derived from constant curvature (lines).
5 CONCLUSIONS

In this study we have extended the FGM method to account for preferential diffusion effects and we validated the approach in DNS of lean premixed methane-hydrogen-air flame kernels. For pure methane flames, preferential diffusion effects may be negligible, but for hydrogen blends they lead to important changes of the flame structure and its dynamics. It was shown that adding one additional degree of freedom to the manifold is enough to capture the main effects for hydrogen blends. This resulted in predictions very close to detailed simulations, but at a fraction of the cost. By adding a stretch term to the flamelet equations, 2D FGM’s can be created that include variations in element mass fractions and enthalpy. At first sight, the exact way how the flame stretch is modeled in the flamelet equations does not appear to be of great importance. Similar results are obtained for two different stretch models. However, different stretch profiles result in different variations in element mass fractions and enthalpy, which may result in different correlations between these changes. If this is the case, than additional degrees of freedom have to be used. How general the correlation between changes in $Z_j$ and $h$ are, and what their influence is on the accuracy of the FGM method, will be the topics of future investigations.

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