

Differential diffusion effects in FGM context for premixed LES of hydrogen blended fuels

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Abstract

The framework of the tabulated chemistry method is used to develop an adequate strategy in order to account for differential diffusion effects without being limited by the necessity of using the detailed chemistry. Therefore, the well established Flamelet Generated Manifolds approach, including detailed information on reaction kinetics and permitting reasonable computational costs, is used within this work. The strong diffusivity of hydrogen blended hydrocarbon fuels significantly alters the flame characteristics and provokes thermo-diffusive instabilities. In order to consider these effects the general transport equations for the reaction progress variable, mixture fraction and enthalpy are extended. The models suitability for LES application is shown by verifying its consistency with laminar unstretched premixed flames obtained by detailed chemistry simulations.

Introduction

In technical applications the combustion of lean premixed hydrogen and hydrogen blended hydrocarbon fuels is taking greater significance not only due to the low emissions characteristics but also to the increasing interest of renewable energy and clean coal technology. The strong diffusivity of the hydrogen substantially alters the flame characteristics like its structure, propagation speed and also the occurrence of thermo-diffusive instabilities. This phenomenon is denoted by differential diffusion where species mass and heat locally diffuse at non-equal rate within the flame [1]. A key parameter representing the molecular diffusion in premixed combustion is the Lewis number, Le , defined as the ratio of thermal to mass diffusivity of the mixtures species. As reported in several experimental [2,3] and numerical [4,5,6] studies, differential diffusion can have important effects in non-premixed and premixed [7] turbulent flames. This goes against the common assumption of negligible molecular transport effects with increasing turbulence intensity. In order to understand from both fundamental and practical perspectives like suggested by Barlow et al. [8] further model development considering differential diffusion effects may be essential for accurate predictions of combustion simulations.

The progressively raising computational resources enable an increasing usage of Large Eddy Simulations (LES) in industrial development processes [9]. However, use of detailed chemistry for industrial applications is still limited. In order to gain detailed chemistry information throughout the flame, a progress variable approach is chosen for this work reducing the computational costs. The Flamelet Generated Manifold (FGM) method [10], which is also referred to as Flame Prolongation of ILDM (FPI) method [11], is based on the general idea mapping all the thermo-chemical scalar information of the combustion to a table from a laminar flamelet database. This manifold is computed in a pre-processing stage from steady adiabatic one-dimensional premixed laminar flamelets with detailed chemistry and

stored into a table depending on a small number of control variables. For every of the independent control variables a transport equation is solved during the simulation. It is well established in the combustion literature that the FGM method has proven to be accurate for various combustion cases [12].

Several modelling strategies for accounting differential diffusion effects in premixed flames have been developed. Some of them account these effects just through the tabulation and filtering of non-unity Lewis number flamelets computed with complex transport models and detailed chemistry [13]. But most of them have in common that for being capable of modelling accurately the combustion of flames with non-unity Lewis number fuels within a reduced chemistry framework new models are necessary with more than a single variable approach. At least a two-equation model is needed for an adequate consideration. These models are based on the progress variable approach being extended with additional transport equations, i.e. for a mixture fraction or enthalpy. Regele et al. [14] are presenting a method based on a two-equation model and on the Flamelet Progress Variable - levelset approach developed by Knudsen and Pitsch [15,16]. They have added a source term to the transport equation of the mixture fraction and assumed a non-unity Lewis number just for the fuel and unity for the rest of the mixture. Another promising method of capturing these effects in context of the FGM method is presented by de Swart et al. [17] and recently by Donini et al. [12]. Both propose a progress variable approach extended with a mixture fraction superposed by weighted element mass fractions. Whereas, the latter one includes a furthermore control variable for the enthalpy. These two methods consider non-unity Lewis numbers for all involved species. Within these approaches a model is derived for the progress variable equation and then generalized for any control variable. In addition the following assumption is made that the control variables are locally a function of the progress variable solely.

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In contrast to the last referred contributions the transport equations for all control variables are not generalized from the progress variable in the present paper. Rather, the differential diffusion effects are accounted in the transport equation of all control variables specifically. Based on first principles effective Schmidt numbers are derived considering non-unity Lewis numbers for all species. As well it is not assumed that the control variables are locally a function of the progress variable exclusively, but rather a function of all control variables.

The main objective of this paper is thus to develop a tabulated chemistry for LES modelling accounting for the complex transport phenomena of differential diffusion. It is structured as follows. After a short introduction into the tabulation procedure the derivation of the evolution equations for the progress variable, mixture fraction and enthalpy is presented. The introduced effective Schmidt numbers within the diffusion term of the filtered scalar transport equations are decomposed into diffusion and cross-diffusion terms. This method is at a first step verified and discussed to be consistent with laminar unstretched premixed flames for several hydrogen mixtures as obtained by detailed chemistry simulations.

The FGM tabulation strategy

In the context of LES the usage of detailed chemistry does still imply too high computational costs in order to provide fully detailed information about the thermo-chemical state throughout the flame. Reduced mechanisms, which are solving only a few transport equations for main species, are more computational feasible but the provided information on minor species is insufficient. The chosen approach is therefore based on the FGM method, following the table generation procedure described by Ketelheun et al. [18]. The FGM is generated in a pre-processing step, where the whole thermo-chemical information is stored into a 3D manifold. This data is gained from one-dimensional laminar premixed flamelet calculations with detailed chemistry and non-unity constant species Lewis number using the laminar flame solver Chem1D [19] coupled with the GRI-Mech 3.0 reaction mechanism [20]. Every flamelet is parameterized by a progress variable Y , describing the reaction progress from unburnt mixture to burnt chemical equilibrium, a mixture fraction Z and the enthalpy h . The latter two control variables are also needed for every flamelet due to the presence of local changes of the mixture fraction and the enthalpy caused by differential diffusion effects. Thus an adapted tabulation procedure is needed mapping correctly the non iso-mixture fraction and non iso-enthalpy flamelet data depending on the control variables. The introduced effective Schmidt numbers for the diffusion and cross-diffusion terms are stored additionally into the same FGM. The complete manifold includes of the species mass fractions Y_k , source terms $\dot{\omega}_k$, density ρ , viscosity μ and other scalar quantities like temperature T or the effective diffusion and cross-diffusion Schmidt numbers $Sc_{CV_j}^{CV_i}$ (see definition below).

$$\Phi = \Phi(Y, Z, h) \text{ with } \Phi \in \{Y_k, \dot{\omega}_k, \rho, \mu, T, Sc_{CV_j}^{CV_i}\} \quad (1)$$

Differential diffusion into the FGM method

The implementation and adaption of this work is realized within the academic code FASTEST, which has a second order accuracy in time and space. The partial differential equations of the incompressible, variable density momentum and continuity are solved within the solver using a pressure correction procedure. Furthermore, the transport equations for all control variables, which are derived in the following subsections are implemented. The transport equations for all control variables are derived from the following evolution equations of the species mass fraction Y_k

$$\rho \frac{DY_k}{Dt} = \frac{\partial \rho Y_k}{\partial t} + \frac{\partial \rho u_i Y_k}{\partial x_i} = \frac{\partial}{\partial x_i} (-j_k) + \dot{\omega}_k \quad (2)$$

where the diffusion velocity j_k can be simplified using Fick's law and Hirschfelder and Curtiss approximation ($-j_k = -Y_k V_k = \rho D_k \frac{\partial Y_k}{\partial x_i}$) [21]. The equivalent diffusion coefficient D_k of species k into the rest of the mixture can also be rewritten as a function of the species Lewis number Le_k , which is a non-dimensional number giving the relation of thermal and mass diffusion, ($\rho D_k = \frac{\lambda}{c_p Le_k}$) leading to

$$\rho \frac{DY_k}{Dt} = \nabla \left(\frac{\lambda}{c_p Le_k} \nabla Y_k \right) + \dot{\omega}_k \quad (3)$$

1. The reaction progress variable equation

The progress variable Y has to be chosen in such a way that a good resolution of the flame is achieved and that it increases monotonically throughout the flame ensuring a unique assignment of the tabulated scalar parameters. The progress variable is defined as N_s linear combinations of a_n weighted species mass fractions Y_n

$$Y = \sum_n^{N_s} a_n Y_n \quad \dot{\omega}_Y = \sum_n^{N_s} a_n \dot{\omega}_n \quad (4)$$

where the source term of the progress variable $\dot{\omega}_Y$ is similarly defined. This leads to the reaction progress variable equation

$$\rho \frac{DY}{Dt} = \nabla \left(\sum_n^{N_s} \frac{\lambda}{c_p Le_n} a_n \nabla Y_n \right) + \dot{\omega}_Y \quad (5)$$

Underlying that the species mass fractions are locally a function of the considered control variables $Y_k = Y_k(Y, Z, h)$, one obtains for the gradients ∇Y_k the relation

$$\nabla Y_k = \frac{\partial Y_k}{\partial Y} \nabla Y \Big|_{Z,h} + \frac{\partial Y_k}{\partial Z} \nabla Z \Big|_{Y,h} + \frac{\partial Y_k}{\partial h} \nabla h \Big|_{Y,Z} \quad (6)$$

which yields the following transport equation

$$\rho \frac{DY}{Dt} = \nabla \left(\frac{\lambda}{c_p} \left(\sum_n^{N_s} \frac{1}{Le_n} a_n \frac{\partial Y_n}{\partial Y} \Big|_{z,h} \right) \nabla Y + \frac{\lambda}{c_p} \left(\sum_n^{N_s} \frac{1}{Le_n} a_n \frac{\partial Y_n}{\partial Z} \Big|_{r,h} \right) \nabla Z + \frac{\lambda}{c_p} \left(\sum_n^{N_s} \frac{1}{Le_n} a_n \frac{\partial Y_n}{\partial h} \Big|_{z,r} \right) \nabla h \right) + \dot{\omega}_Y \quad (7)$$

The diffusion term on the right hand side (RHS) now consists of inner diffusion and cross-diffusion terms, which are locally dependent on the gradients of all control variables (Y, Z and h).

Rewriting the reaction progress variable equation by describing the coefficients of the diffusion terms one obtains

$$\rho \frac{DY}{Dt} = \nabla \left(\frac{\mu}{Sc_Y^Y} \nabla Y \right) + \nabla \left(\frac{\mu}{Sc_Z^Y} \nabla Z \right) + \nabla \left(\frac{\mu}{Sc_h^Y} \nabla h \right) + \dot{\omega}_Y \quad (8)$$

where the effective Schmidt numbers are defined as

$$Sc_Y^Y = \frac{\mu c_p}{\lambda} \left(\sum_n^{N_s} \frac{1}{Le_n} a_n \frac{\partial Y_n}{\partial Y} \Big|_{z,h} \right)^{-1} \quad (9)$$

$$Sc_Z^Y = \frac{\mu c_p}{\lambda} \left(\sum_n^{N_s} \frac{1}{Le_n} a_n \frac{\partial Y_n}{\partial Z} \Big|_{r,h} \right)^{-1} \quad (10)$$

$$Sc_h^Y = \frac{\mu c_p}{\lambda} \left(\sum_n^{N_s} \frac{1}{Le_n} a_n \frac{\partial Y_n}{\partial h} \Big|_{z,r} \right)^{-1} \quad (11)$$

2. The mixture fraction equation

The mixture fraction is a dimensionless number describing the ratio of fuel to oxidizer within the mixture. There are many definitions for it, all based on linear relations to the element mass fractions Z_j , which defines the ratio between the mass of an element j and the total mass,

$$Z_j = \sum_k^{N_{Sp}} \mu_{jk} Y_k = \sum_k^{N_{Sp}} a_{jk} \frac{M_j}{M_k} Y_k \quad (12)$$

where N_{Sp} denotes the total number of all species involved within the reaction mechanism, μ_{jk} the mass proportion and a_{jk} the number of atoms of the element j in the species k and M_i the element/species mass.

Based on (3) and the N_{El} linear combinations of the γ_j weighted element mass fractions Z_j the non-normalized mixture fraction Z^* reads

$$Z^* = \sum_j^{N_{El}} \gamma_j Z_j = \sum_j^{N_{El}} \gamma_j \sum_k^{N_{Sp}} a_{jk} \frac{M_j}{M_k} Y_k \quad (13)$$

Normalizing it by using values for pure fuel and oxidizer one obtains for the normalized mixture fraction Z

$$Z = \frac{Z^* - Z_{Ox}^*}{Z_{Fu}^* - Z_{Ox}^*} \quad (14)$$

$$Z = \frac{\sum_j^{N_{El}} \gamma_j \sum_k^{N_{Sp}} a_{jk} \frac{M_j}{M_k} (Y_k - Y_{k,Ox})}{\sum_j^{N_{El}} \gamma_j \sum_k^{N_{Sp}} a_{jk} \frac{M_j}{M_k} (Y_{k,Fu} - Y_{k,Ox})} \quad (15)$$

where $C = \sum_j^{N_{El}} \gamma_j \sum_k^{N_{Sp}} a_{jk} \frac{M_j}{M_k} (Y_{k,Fu} - Y_{k,Ox})$ is a constant denominator. The conservation equation of the mixture fraction thus results in:

$$\rho \frac{DZ}{Dt} = \nabla \left(\frac{1}{C} \sum_j^{N_{El}} \gamma_j \sum_k^{N_{Sp}} a_{jk} \frac{M_j}{M_k} \frac{\lambda}{c_p} \frac{1}{Le_k} \nabla Y_k \right) \quad (16)$$

where the sum of all sources ($\sum_k^{N_{Sp}} \dot{\omega}_k = 0$) and the derivation of the constant species mass fractions in the oxidizer ($\nabla Y_{k,Ox} = 0$) vanishes. In case of using Bilger's definition or the ones used within this work the weighting factors γ_j are listed in Table 1.

Table 1
Weighting factors of the mixture fractions definition

	$j = C$	$j = H$	$j = O$	$j = N$	$j = Ar$
$\gamma_{j,Bilger}$	$\frac{2}{M_C}$	$\frac{1}{M_H}$	$-\frac{1}{M_O}$	0	0
γ_j	1	1	0	0	0

Utilizing (6) and rewriting the conservation equation as

$$\rho \frac{DZ}{Dt} = \nabla \left(\frac{\mu}{Sc_Y^Z} \nabla Y \right) + \nabla \left(\frac{\mu}{Sc_Z^Z} \nabla Z \right) + \nabla \left(\frac{\mu}{Sc_h^Z} \nabla h \right) \quad (17)$$

the effective diffusion and cross-diffusion Schmidt numbers for the mixture fraction equations are

$$Sc_Y^Z = C \frac{\mu c_p}{\lambda} \left(\sum_j^{N_{El}} \gamma_j \sum_k^{N_{Sp}} a_{jk} \frac{M_j}{M_k} \frac{1}{Le_k} \frac{\partial Y_k}{\partial Y} \Big|_{z,h} \right)^{-1} \quad (18)$$

$$Sc_Z^Z = C \frac{\mu c_p}{\lambda} \left(\sum_j^{N_{El}} \gamma_j \sum_k^{N_{Sp}} a_{jk} \frac{M_j}{M_k} \frac{1}{Le_k} \frac{\partial Y_k}{\partial Z} \Big|_{r,h} \right)^{-1} \quad (19)$$

$$Sc_h^Z = C \frac{\mu c_p}{\lambda} \left(\sum_j^{N_{El}} \gamma_j \sum_k^{N_{Sp}} a_{jk} \frac{M_j}{M_k} \frac{1}{Le_k} \frac{\partial Y_k}{\partial h} \Big|_{r,z} \right)^{-1} \quad (20)$$

3. The enthalpy equation

The addition of the enthalpy as a third table dimension is considered not only due to a possible consideration of heat transfer at walls or by radiation but also due to changes of enthalpy throughout a flame front caused by differential diffusion effects. The enthalpy is defined as the sum of the sensible and chemical enthalpy and reads

$$h = \sum_k^{N_{Sp}} h_k Y_k \quad h_k = \int_{T_0}^T c_{p,k} dT + \Delta h_{f,k}^0 \quad (21)$$

where h_k , $c_{p,k}$ and $\Delta h_{f,k}^0$ are respectively the enthalpy, the specific heat capacity at constant pressure and the enthalpy formation at the reference temperature T_0 of species k . The general transport equation for the enthalpy [22] is given by

$$\rho \frac{Dh}{Dt} = \frac{Dp}{Dt} + \tau_{ij} \frac{\partial u_j}{\partial x_i} + \dot{Q} + \rho \sum_k^{N_{Sp}} Y_k f_{k,i} V_{k,i} - \frac{\partial \dot{q}_i}{\partial x_i} \quad (22)$$

This equation can be simplified by disregarding the first two terms on the RHS due to the low Mach number assumption ($Ma < 0.3$), the third term \dot{Q} due to missing heat sources and sinks within the computational domain and the fourth term which includes volume forces $f_{k,i}$ due to the sole consideration of the constant appearing gravity equally on all species. The last remaining term is the enthalpy flux \dot{q}_i due to heat conduction and species diffusion with different enthalpies ($\dot{q}_i = -\lambda \nabla T + \rho \sum_k^{N_{Sp}} h_k Y_k V_{k,i}$). This can also be transformed using $dh = c_p dT + \sum_k^{N_{Sp}} h_k dY_k$ [23] and the same approximation as used in (3) to the following expression

$$\dot{q}_i = -\frac{\lambda}{c_p} \nabla h + \frac{\lambda}{c_p} \sum_k^{N_{Sp}} h_k \left(1 - \frac{1}{Le_k}\right) \nabla Y_k \quad (23)$$

The simplified and approximated enthalpy equation

combined with (6) and considering differential diffusion effects thus results in

$$\rho \frac{Dh}{Dt} = \nabla \left(\frac{\mu}{Pr_Y} \nabla Y \right) + \nabla \left(\frac{\mu}{Pr_Z} \nabla Z \right) + \nabla \left(\frac{\mu}{Pr_h} \nabla h \right) \quad (24)$$

where the effective differential diffusion Prandtl numbers are defined as

$$Pr_Y = \frac{\mu c_p}{\lambda} \left(\sum_k^{N_{Sp}} h_k \left(\frac{1}{Le_k} - 1 \right) \frac{\partial Y_k}{\partial Y} \Big|_{Z,h} \right) \quad (25)$$

$$Pr_Z = \frac{\mu c_p}{\lambda} \left(\sum_k^{N_{Sp}} h_k \left(\frac{1}{Le_k} - 1 \right) \frac{\partial Y_k}{\partial Z} \Big|_{Y,h} \right) \quad (26)$$

$$Pr_h = \frac{\mu c_p}{\lambda} \left(1 + \sum_k^{N_{Sp}} h_k \left(\frac{1}{Le_k} - 1 \right) \frac{\partial Y_k}{\partial h} \Big|_{Y,Z} \right) \quad (27)$$

Verification

In order to verify the models suitability for LES applications and further extensive investigations, various unstretched laminar premixed flames were computed in FASTEST and compared to results gained from detailed chemistry calculations with specific constant Lewis numbers performed with Chem1D and GRI Mech 3.0. The boundary conditions for these verification cases were varied in mixture fraction and composition of the hydrogen blended hydrocarbon fuels. **Figure 1** shows the profiles of density, temperature, progress variable and mixture fraction of one-dimensional laminar FASTEST and Chem1D simulations representatively for a lean and rich mixture fraction $Z = \{0.045, 0.055\}$ and two mole fractions of hydrogen $X_{H_2} = \{0.0, 0.4\}$.

The results obtained by considering the differential diffusion effects or not, with respectively unity and non-unity Lewis numbers, show already deviations within detailed chemistry results also for pure methane mixtures, even though the fuel-air mixtures are predominantly consisting of less diffusive species. As expected

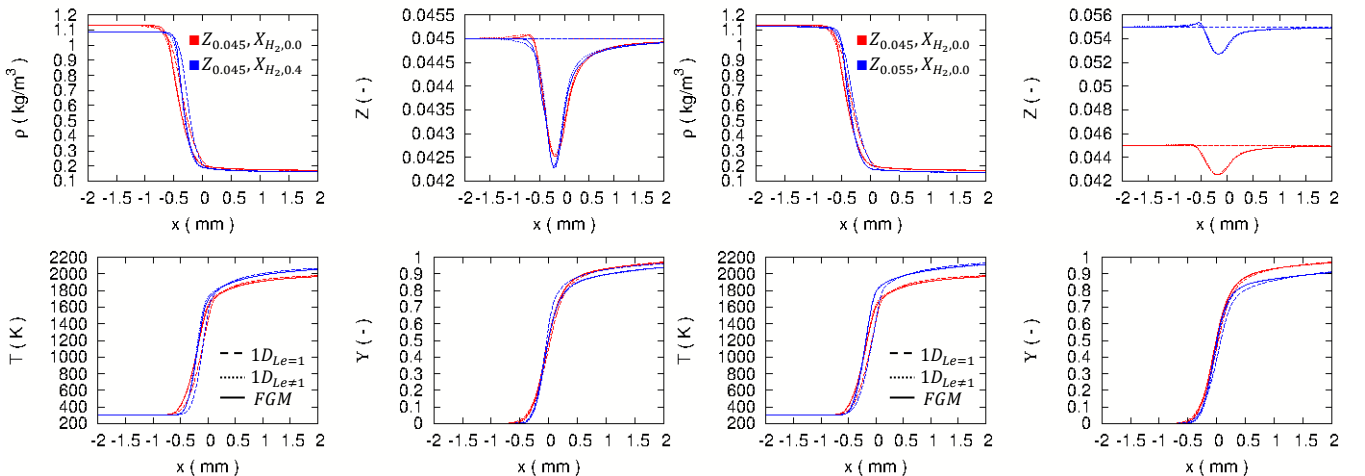


Figure 1 Profiles of density, temperature, mixture fraction and progress variable, where red lines are results for $Z = 0.045$ $X_{H_2} = 0.0$, blue lines at the left are for $Z = 0.045$ $X_{H_2} = 0.4$ and at right for $Z = 0.055$ $X_{H_2} = 0.0$. The full lines (— FGM) are results from the FGM simulations, whereas the dashed (--- $Le = 1$) and dotted (..... $Le \neq 1$) from detailed chemistry.

the main deviation is seen in the mixture fraction profile, where it is not constant through the flame front in the case the differential diffusion effects are neglected. Obviously the mixture fraction shows non-negligible variations within the flame already for less diffusive fuels which are growing for more diffusive hydrogen blended fuels. The reason for this may base on the formation of more diffusive species during the combustion process. Nevertheless, the extended FGM model delivers significantly good results representing the existent diffusion effects.

Conclusions

The consideration of the differential diffusion effects in a three-dimensional manifold covering all thermochemical states within the well-established FGM method shows that additional effective diffusion and cross-diffusion Schmidt and Prandtl numbers appear into the manifold. This is generated from a variety of flamelets with diverse mixture fractions and enthalpies. The models functionality is demonstrated by integrating it into the three-dimensional LES code FASTEST. Several hydrogen mixtures are used to demonstrate the model's ability to accurately capture the effects of differential diffusion. The model is verified to be consistent with laminar unstretched premixed flames as obtained by detailed chemistry simulations. The correct prediction of important properties for the accurate consideration of the differential diffusion effects, like the mixture fraction variation throughout the flame, by only a few control variables shows the models suitability for LES applications. Further investigations need to be carried out for a detailed assessment of the modeling presented in this paper.

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