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A flamelet model for premixed stretched flames

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Abstract. A mass-based stretch rate is introduced. Combining this stretch rate with the conservation equations for premixed flames gives a set of quasi-one-dimensional flamelet equations, which describe the structure and the mass burning rate of these flames. An integral analysis is applied to the flamelet equations and subsequently a general expression for the mass burning rate of stretched flames is derived. Finally, the mass burning rate is computed for weakly stretched methane/air flames.

1 Introduction

Research on stretched flames was initiated by Karlovitz et al. [11], Lewis and von Elbe [13] and Markstein [17]. Since these early publications, a lot of papers on stretched flames have been published. Many of these papers are based on large activation energy asymptotics; see e.g. [2, 18]. A different approach is the integral analysis by Chung and Law [4]. Analytical studies of stretched flames are often based on simple models, with e.g. a flame sheet, a single irreversible reaction and a single Lewis number. Stimulated by the ideas of Buckmaster [2], who first realized that the flame structure in the preheat zone is of importance and using the ideas of the integral analysis of Chung and Law [4], we recently extended the flame stretch theory to more general flames, with finite flame front thickness [8, 9].

The stretch rate, as introduced by Karlovitz et al.[11], is defined as the fractional rate of change of an infinitesimal small area on a flame sheet due to the movement of this sheet. This stretch definition is based on the flame sheet model and does not take into account the internal structure of a flame. In [9], we have introduced a mass-based definition of the stretch rate, which is applicable to general three-dimensional, instationary flames with finite flame front thickness. More specifically, we define the stretch rate as the fractional rate of change of mass in an infinitesimal small volume in the flame front, due to the movement of the flame front. For the flame sheet model, our definition reduces to the classical definition.

Substitution of the new stretch rate definition into the continuity equation gives a quasi-one-dimensional equation, describing mass transport perpendicular to the flame front. Likewise, introducing the stretch rate into the combustion equations, these equations reduce to a set of quasi-one-dimensional conservation equations, describing transport perpendicular to the flame front. Transport terms in the flame front are included in source terms, which are proportional to the stretch rate. These equations describe the inner structure and the mass burning rate of the flame front and hold in the flamelet regime, when the characteristic length of perturbations of the flame front is much larger than the flame thickness. Therefore, these equations are referred to as the 'flamelet' equations. The concept of flamelets is introduced for turbulent flames; see e.g. [19]. However, in this paper we apply the flamelet theory to laminar flames. We like to emphasize that the flamelet equations are quite generally valid, for flames of finite thickness, for any number of chemical species etc. The propagation of a flame, for given mass burning rate, is determined by the G -equation and the momentum equations. This means that the flamelet equations together with the momentum equations and the G -equation constitute a suitable model for the analysis of laminar, premixed flames. In this paper, we focus on the flamelet equations, i.e. for a given stretch rate we investigate the flamelet equations. In particular, we study the influence of flame stretch on the mass burning rate.

We have organized this paper as follows. In the next section, we present a mathematical model for laminar, premixed flames, including the conservation equations for mass, species mass fractions, enthalpy and element mass fractions. Then, in Section 3, we introduce a mass-based definition of flame stretch and derive the flamelet equations. In Section 4, we apply the integral analysis to the flamelet equations and derive a general expression for the mass burning rate, which depends on the enthalpy and element composition in the burnt gas. We also derive integral balances for the enthalpy and element mass fractions. In Section 5, we explicitly compute the mass burning rate from this algebraic model for the special case of lean methane/air flames.

2 Governing equations

In this section we present a mathematical model for laminar, premixed flames under atmospheric conditions. Typically, we consider the combustion of hydrocarbons in air. The species in these flames are numbered 1 through N_s . Species N_s is nitrogen (N_2) and is present in abundance, while the other species are considered as trace species. For a *given* flow field with velocity \mathbf{v} and pressure p , the conservation equations for the mass density ρ , the first $N_s - 1$ species mass fractions Y_i and the specific enthalpy h read [14, 24]:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = 0, \quad (1)$$

$$\frac{\partial (\rho Y_i)}{\partial t} + \nabla \cdot (\rho \mathbf{v} Y_i) + \nabla \cdot (\rho Y_i \mathbf{V}_i) = w_i, \quad i = 1, \dots, N_s - 1, \quad (2)$$

$$\frac{\partial (\rho h)}{\partial t} + \nabla \cdot (\rho \mathbf{v} h) = -\nabla \cdot \mathbf{q}, \quad (3)$$

where \mathbf{V}_i and w_i are the diffusion velocity and the reaction rate of species i , respectively, and where \mathbf{q} is the heat flux vector. The mass fraction Y_{N_2} follows from the requirement that all mass fractions add up to one, and thus we do not have to solve a conservation equation as in (2) for nitrogen. The conservation equations (1)-(3) have to be completed with the caloric equation of state

$$h = \sum_{i=1}^{N_s} Y_i h_i \quad \text{with} \quad h_i = h_i^0 + \int_{T_0}^T c_{p,i}(\vartheta) d\vartheta, \quad (4)$$

which defines h as a function of the temperature T and the species mass fractions Y_i , and the thermal equation of state

$$p_{amb} = \frac{\rho RT}{M} \quad \text{with} \quad \frac{1}{M} = \sum_{i=1}^{N_s} \frac{Y_i}{M_i}. \quad (5)$$

In equation (4), h_i , h_i^0 and $c_{p,i}$ are the specific enthalpy, the specific enthalpy of formation at reference temperature T_0 and the specific heat at constant pressure of species i , respectively. In equation (5), R is the universal gas constant, M is the average molar mass and M_i is the molar mass of species i . The pressure p is set to a constant value p_{amb} in (5), which is valid for low Mach number flow. In this paper, we study stretched flames in a given flow field on the basis of equations (1)-(5). Conversely, we do not investigate the influence of these flames on the flow, and therefore we do not consider the momentum equations. Stated otherwise, the modelling of a laminar flame can be decoupled in a combustion part, presented in this section, and a flow part, given by the momentum equations. This approach does not presuppose a passive flow field; flame and flow do influence each other. However, in this paper we solely consider the effect of the flow on the flame.

Models for the reaction rates w_i are reported in literature; see e.g. [23, 24]. For the diffusion velocities \mathbf{V}_i of the trace species we employ the so-called generalized law of Fick [16, 20], i.e.

$$Y_i \mathbf{V}_i = -D_{i,m} \nabla Y_i, \quad i = 1, \dots, N_s - 1, \quad (6)$$

where $D_{i,m}$ is the mixture-averaged diffusion coefficient, describing the diffusivity of species i in the mixture. Besides, the diffusion velocity of nitrogen follows from the constraint

$$\sum_{i=1}^{N_s} Y_i \mathbf{V}_i = \mathbf{0}, \quad (7)$$

stating that there is no overall diffusion of mass. For the heat flux vector we use the common expression [24]

$$\mathbf{q} = -\lambda \nabla T + \rho \sum_{i=1}^{N_s} h_i Y_i \mathbf{V}_i, \quad (8)$$

where λ is the thermal conductivity. This expression for \mathbf{q} only takes into account enthalpy transport through conduction and mass diffusion. Using equations (4), (6), (7) and (8), the conservation equations for Y_i and h can be rewritten as:

$$\frac{\partial(\rho Y_i)}{\partial t} + \nabla \cdot (\rho \mathbf{v} Y_i) - \frac{1}{Le_i} \nabla \cdot \left(\frac{\lambda}{c_p} \nabla Y_i \right) = w_i, \quad i = 1, \dots, N_s - 1, \quad (9)$$

$$\frac{\partial(\rho h)}{\partial t} + \nabla \cdot (\rho \mathbf{v} h) - \nabla \cdot \left(\frac{\lambda}{c_p} \nabla h \right) = \sum_{i=1}^{N_s-1} \left(\frac{1}{Le_i} - 1 \right) \nabla \cdot \left(\frac{\lambda}{c_p} h_i^* \nabla Y_i \right), \quad (10)$$

where $c_p = \sum_{i=1}^{N_s} Y_i c_{p,i}$ is the specific heat at constant pressure of the mixture and where $h_i^* = h_i - h_{N_2}$ ($i = 1, \dots, N_s - 1$). The right hand side of Eq. (10) describes enthalpy transport of the trace species relative to the enthalpy of the inert N_2 . In these equations we introduced the Lewis numbers $Le_i = \lambda / (\rho D_{i,m} c_p)$ ($i = 1, \dots, N_s - 1$), which we assume to be constant.

Alternatively, the enthalpy conservation equation (10) can be formulated in terms of the thermal enthalpy h_T , which is defined by

$$h_T = \int_{T_0}^T c_p(\vartheta, Y_i) d\vartheta. \quad (11)$$

Combining equations (9) and (10) and using the relation

$$h = h_T + \sum_{i=1}^{N_s-1} Y_i h_i^0, \quad (12)$$

which follows readily from equations (4) and (11), we obtain the following conservation equation for h_T :

$$\frac{\partial(\rho h_T)}{\partial t} + \nabla \cdot (\rho \mathbf{v} h_T) - \nabla \cdot \left(\frac{\lambda}{c_p} \nabla h_T \right) = \sum_{i=1}^{N_s-1} \left(\frac{1}{Le_i} - 1 \right) \nabla \cdot \left(\frac{\lambda}{c_p} h_{T,i}^* \nabla Y_i \right) + w_{h_T}, \quad (13)$$

where $h_{T,i}^* = h_{T,i} - h_{T,N_2}$ ($i = 1, \dots, N_s - 1$) and where $w_{h_T} = -\sum_{i=1}^{N_s-1} h_i^0 w_i$. The first term in the right-hand side of equation (13), which describes transport of thermal enthalpy relative to the thermal enthalpy of N_2 , is usually small and even vanishes in case $c_{p,i} = c_p$ for all species i . Summarizing, we have the conservation equations (1), (9) and (10) for ρ , Y_i and h or, alternatively, (1), (9) and (13) for ρ , Y_i and h_T .

In the following of this paper, we need the element mass fractions Z_j for the computation of the mass burning rate. Assume that the elements are numbered 1 through N_e , and that the N_e th element is nitrogen (N). The element mass fractions Z_j of the other elements are defined by

$$Z_j = \sum_{i=1}^{N_s-1} w_{j,i} Y_i, \quad j = 1, \dots, N_e - 1, \quad (14)$$

where $w_{j,i}$ is the mass fraction of element j in species i , i.e. $w_{j,i} = W_j \mu_{j,i} / M_i$ with W_j the molar mass of element j and with $\mu_{j,i}$ the number of atoms of element j in species i . Taking the proper linear combination of the species equations (9), we get the following conservation equations for Z_j :

$$\frac{\partial(\rho Z_j)}{\partial t} + \nabla \cdot (\rho \mathbf{v} Z_j) - \nabla \cdot \left(\frac{\lambda}{c_p} \nabla Z_j \right) = \sum_{i=1}^{N_s-1} \left(\frac{1}{Le_i} - 1 \right) w_{j,i} \nabla \cdot \left(\frac{\lambda}{c_p} \nabla Y_i \right),$$

$$j = 1, \dots, N_e - 1. \quad (15)$$

These $N_e - 1$ conservation equations are, of course, *not* independent of the set (9). Note the similarity between the conservation equations (10) and (15) for h and Z_j , respectively. These equations are all of convection-diffusion type with a source term proportional to the mass diffusion fluxes of the trace species.

3 Flamelet equations

In this section we introduce a mass-based definition of flame stretch and combine this definition with the conservation equations of the previous section. As a result we obtain a quasi-one-dimensional set of equations, referred to as the flamelet equations, which describe the internal structure and the mass burning rate of a flame. The flame stretch formalism presented in [8, 9] will be the starting point of this section. The reader is referred to these papers for more details.

A premixed flame is defined as the region in space, where a scalar variable G assumes values between the unburnt (G_u) and burnt (G_b) values. We assume that $\nabla G \neq \mathbf{0}$ everywhere in the flame. The variable G can be, for instance, one of the species mass fractions or the thermal enthalpy, and will be specified later. A 'flame surface' is defined as an iso-plane of G , i.e. a surface where $G(\mathbf{x}, t) = G_0$ for arbitrary constant G_0 . The motion of such a surface is described by the kinematic condition

$$\frac{dG}{dt} := \frac{\partial G}{\partial t} + (\mathbf{v}_f \cdot \nabla) G = 0, \quad (16)$$

stating that a point on a flame surface stays on this surface for all t . Here, \mathbf{v}_f is the local velocity of a flame surface. A local orthogonal coordinate system $\boldsymbol{\eta} = (\xi, \zeta, \eta)$ is defined in terms of these flame surfaces, with (ξ, ζ) an orthogonal coordinate system in each flame surface and η the coordinate perpendicular to the flame surfaces; see Fig. 1. The factors $h_\xi = \left| \frac{\partial \mathbf{x}}{\partial \xi} \right|$, $h_\zeta = \left| \frac{\partial \mathbf{x}}{\partial \zeta} \right|$ and $h_\eta = \left| \frac{\partial \mathbf{x}}{\partial \eta} \right|$ are scale factors, defining the arc-lengths in the $\boldsymbol{\eta}$ -coordinate system. Note that the unit normal vector \mathbf{e}_η on a flame surface can be written as $\mathbf{e}_\eta = \text{sign}(G_b - G_u) \nabla G / |\nabla G|$, so that \mathbf{e}_η is directed to the burnt gas mixture.

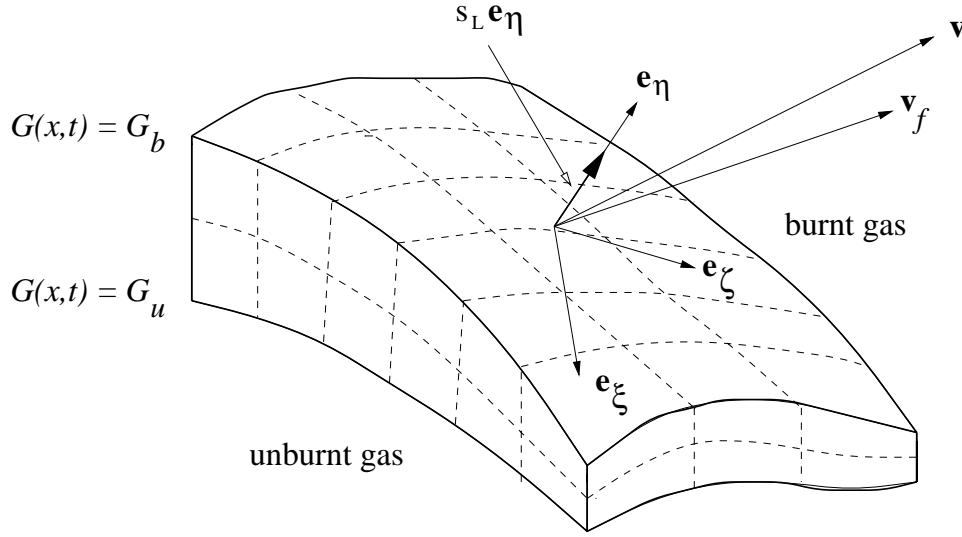


Figure 1: Curvilinear coordinate system attached to the flame.

We define the stretch rate field K in the flame as the fractional rate of change of the mass $M(t)$ contained in an infinitesimal volume $V(t)$ in the flame, moving with velocity \mathbf{v}_f [9]:

$$K = \frac{1}{M} \frac{dM}{dt} \quad \text{with} \quad M(t) = \int_{V(t)} \rho \, dV. \quad (17)$$

Applying the transport theorem [1] to $M(t)$ gives the following expression for K :

$$\rho K = \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}_f). \quad (18)$$

Apart from the usual terms related to flame curvature and flow straining, this definition of K incorporates additional contributions, for instance due to flame thickness variations.

Combining equation (18) with the continuity equation (1) gives

$$\nabla \cdot (\rho(\mathbf{v} - \mathbf{v}_f)) = -\rho K. \quad (19)$$

Taking into account that the tangential component $\mathbf{v}_{f,t}$ of the flame surface velocity is equal to the tangential component \mathbf{v}_t of the fluid velocity, this implies that all deviations from local one-dimensional flow are gathered in the stretch rate K . Also note that the difference $\rho(\mathbf{v} - \mathbf{v}_f)$ is equal to the amount of mass effectively consumed by the flame, so that we may write

$$\rho(\mathbf{v} - \mathbf{v}_f) = m \mathbf{e}_\eta \quad \text{with} \quad m = \rho s_L, \quad (20)$$

where s_L is the local burning velocity and is m the mass burning rate. Substitution of (20) into (19) then gives the quasi-one-dimensional continuity equation

$$\frac{\partial}{\partial s} (\sigma m) = -\sigma \rho K, \quad (21)$$

where the coordinate s in the flame is defined by $ds = h_\eta d\eta$ and where the variable $\sigma = h_\xi h_\zeta = \left| \frac{\partial \mathbf{x}}{\partial \xi} \times \frac{\partial \mathbf{x}}{\partial \zeta} \right|$ is a measure for the area on the flame surfaces. The variable σm is thus the mass flow rate through the flame surfaces and the source term $-\sigma \rho K$ describes mass transport in these surfaces.

Our definition of the stretch rate can be justified further by considering the conservation equation for G :

$$\frac{\partial(\rho G)}{\partial t} + \nabla \cdot (\rho \mathbf{v} G) - \frac{1}{Le_G} \nabla \cdot \left(\frac{\lambda}{c_p} \nabla G \right) = w_G, \quad (22)$$

where Le_G and w_G are the Lewis number and the source term of variable G , respectively. If $G = h$ or $G = h_T$ then obviously $Le_G = 1$. Using the kinematic condition (16) and equations (18) and (20), the conservation equation for G can be written in the quasi-one-dimensional form

$$\frac{\partial}{\partial s}(\sigma m G) - \frac{1}{Le_G} \frac{\partial}{\partial s} \left(\sigma \frac{\lambda}{c_p} \frac{\partial G}{\partial s} \right) - \sigma w_G = -\sigma \rho K G, \quad (23)$$

with all transport terms in the flame surfaces gathered in the right-hand side term $-\sigma \rho K G$.

The same procedure can be applied to the conservation equations of the other scalar variables Y_i , h_T , h and Z_j , and leads to the following set of equations:

$$\begin{aligned} \frac{\partial}{\partial s}(\sigma m) &= -\sigma \rho K, \\ \frac{\partial}{\partial s}(\sigma m Y_i) - \frac{1}{Le_i} \frac{\partial}{\partial s} \left(\sigma \frac{\lambda}{c_p} \frac{\partial Y_i}{\partial s} \right) - \sigma w_i &= -\sigma \rho K Y_i + Q_{Y_i}, \quad i = 1, \dots, N_s - 1, \\ \frac{\partial}{\partial s}(\sigma m h_T) - \frac{\partial}{\partial s} \left(\sigma \frac{\lambda}{c_p} \frac{\partial h_T}{\partial s} \right) - \sum_{i=1}^{N_s-1} \left(\frac{1}{Le_i} - 1 \right) \frac{\partial}{\partial s} \left(\sigma \frac{\lambda}{c_p} h_{T,i}^* \frac{\partial Y_i}{\partial s} \right) - \sigma w_{h_T} &= \\ &= -\sigma \rho K h_T + Q_{h_T}, \\ \frac{\partial}{\partial s}(\sigma m h) - \frac{\partial}{\partial s} \left(\sigma \frac{\lambda}{c_p} \frac{\partial h}{\partial s} \right) - \sum_{i=1}^{N_s-1} \left(\frac{1}{Le_i} - 1 \right) \frac{\partial}{\partial s} \left(\sigma \frac{\lambda}{c_p} h_i^* \frac{\partial Y_i}{\partial s} \right) &= -\sigma \rho K h + Q_h, \\ \frac{\partial}{\partial s}(\sigma m Z_j) - \frac{\partial}{\partial s} \left(\sigma \frac{\lambda}{c_p} \frac{\partial Z_j}{\partial s} \right) - \sum_{i=1}^{N_s-1} \left(\frac{1}{Le_i} - 1 \right) w_{j,i} \frac{\partial}{\partial s} \left(\sigma \frac{\lambda}{c_p} \frac{\partial Y_i}{\partial s} \right) &= \\ &= -\sigma \rho K Z_j + Q_{Z_j}, \quad j = 1, \dots, N_e - 1, \quad (24) \end{aligned}$$

where the terms Q_{Y_i} , Q_{h_T} , Q_h and Q_{Z_j} , which describe transport in the flame surfaces, arise because the local iso-surfaces of the variables Y_i , h_T , h and Z_j generally do not coincide with the iso-surfaces of G . In the flamelet regime we consider, the characteristic length scale of

perturbations of the flame front is much larger than its thickness, and consequently these terms are small compared to the stretch terms in the right-hand side of (24) and will therefore be omitted in the following. This approximation is equivalent with the assumption that the thermal enthalpy h_T and the mass fractions Y_i ($i = 1, \dots, N_s - 1$) of the trace species are all constant in the flame surfaces. We refer to the equations in (24) as the 'flamelet' equations. We like to emphasize that the set of flamelet equations (24) is quite generally valid, since it is based on the flame model presented in Section 2. In particular, the flamelet equations hold for 3D, time dependent flames of finite thickness, with multiple-species transport and chemistry. The only restriction is that the flamelet approximation should hold, i.e. the Q -terms in (24) are negligible.

The flamelet equations (24) describe the inner structure and the mass burning rate of the flame. Clearly, there is a coupling between the flame and the flow. The influence of the flame on the flow is mainly determined by the density $\rho(s)$. Conversely, the influence of the flow on the flame can be quantified by the stretch rate $K(s)$ and the flame area surface $\sigma(s)$. For given stretch rate $K(s)$ and flame surface area $\sigma(s)$, the combustion variables and the mass burning rate can be computed numerically from the flamelet equations (24). On the other hand, propagation of a flame is determined by the momentum equations and the G -equation [19]

$$\rho \left(\frac{\partial G}{\partial t} + \mathbf{v} \cdot \nabla G \right) = m |\nabla G|, \quad (25)$$

which we can easily derive from equations (16) and (20). Solving the latter equations, using the mass burning rate $m(s)$ computed from (24), gives the new flame configuration, i.e. $K(s)$ and $\sigma(s)$ in the flame. Solution methods for the G -equation are discussed in e.g. [21, 22]. Subsequently, we have to solve the flamelet equations, etc. This approach requires the numerical solution of the flamelet equations for many different fields $K(s)$ and $\sigma(s)$, and can be rather laborious. Instead, we will derive an approximate expression for the mass burning rate, that circumvents the numerical solution of the flamelet equations, based on an integral analysis of these equations. This way, we obtain an efficient flamelet model for laminar flames, which will be elaborated in the next section.

4 Integral analysis of the flamelet equations

In order to compute the mass burning rate, we first derive relations between the state variables $\boldsymbol{\psi} = (h, Z_1, \dots, Z_{N_e-1})$ in the burnt and unburnt gas mixtures by integrating the corresponding flamelet equations across the flame. Subsequently, we derive a general expression for the mass burning rate in the burnt gas from the flamelet equation for G .

We first consider stretchless flames, i.e. $K(s) = 0$. Integration of the flamelet equations for mass, enthalpy and element mass fractions of the set (24) from the unburnt to the burnt gas mixture gives

$$\begin{aligned} (\sigma m^0)_b &= (\sigma m^0)_u, \\ (\sigma m^0 h^0)_b &= (\sigma m^0 h^0)_u, \end{aligned}$$

$$(\sigma m^0 Z_j^0)_b = (\sigma m^0 Z_j^0)_u, \quad j = 1, \dots, N_e - 1, \quad (26)$$

where we have assumed that the conductive and all diffusive fluxes vanish at the unburnt ($s = s_u$) and burnt ($s = s_b$) side of the flame. The superscript '0' refers to the stretchless solution of the flamelet equations and the subscripts 'b' and 'u' denote the values of a variable in the burnt and unburnt gas, respectively. From (26) we can easily see that $h_b^0 = h_u^0$ and $Z_{j,b}^0 = Z_{j,u}^0$, i.e. the enthalpy and the element composition are unchanged after combustion.

Next, we want to compute the stretchless mass burning rate m_b^0 in the burnt gas. Consider equation (23) for G with $K = 0$, which can be compactly written as

$$\mathcal{C}(s) - \frac{\partial}{\partial s} \mathcal{D}(s) = \mathcal{S}(s), \quad (27)$$

where \mathcal{C} , \mathcal{D} and \mathcal{S} are defined by

$$\mathcal{C}(s) = \frac{\partial}{\partial s} (\sigma m^0 G^0), \quad \mathcal{D}(s) = \frac{1}{Le_G} \sigma \frac{\lambda}{c_p} \frac{\partial G^0}{\partial s}, \quad \mathcal{S}(s) = \sigma w_G. \quad (28)$$

We multiply equation (27) with $\mathcal{D}(s)$ and subsequently integrate the resulting equation over s from s_u to s_b . The integral over the diffusion term $\frac{1}{2} \frac{\partial \mathcal{D}^2}{\partial s}$ then drops out, as the diffusion fluxes are zero in the (un)burnt mixture and we find

$$\int_{s_u}^{s_b} \mathcal{C}(s) \mathcal{D}(s) ds = \int_{s_u}^{s_b} \mathcal{S}(s) \mathcal{D}(s) ds. \quad (29)$$

The term $\mathcal{D}(s)$ in the left-hand side of this equation can be obtained by integrating equation (27) over the preheat zone, where $|\mathcal{S}| \ll |\mathcal{C}|$. This results in the approximation

$$\int_{s_u}^{s_b} \mathcal{C}(s) \left(\int_{s_u}^s \mathcal{C}(t) dt \right) ds = \frac{1}{2} \left(\int_{s_u}^{s_b} \mathcal{C}(s) ds \right)^2 \approx \int_{s_u}^{s_b} \mathcal{S}(s) \mathcal{D}(s) ds. \quad (30)$$

This approximation of $\mathcal{D}(s)$ is justified since the reaction zone is much thinner than the preheat zone, so that the major contribution to the integral of $\mathcal{C}\mathcal{D}$ in equation (29) comes from the preheat zone. It should be noted that equation (30) is exact when the reaction layer thickness goes to zero for infinite activation energy. Our approach is then equivalent to large activation energy asymptotics to compute the mass burning rate [3].

Evaluation of the integrals in (30) and using $(\sigma m^0)_b = (\sigma m^0)_u$ gives the following approximation for the mass burning rate:

$$m_b^0 \approx \mathcal{F}(\boldsymbol{\psi}_b^0; G_u^0), \quad (31)$$

where the function $\mathcal{F}(\boldsymbol{\psi}_b; G_u)$ is defined by

$$\mathcal{F}(\boldsymbol{\psi}_b; G_u) = \frac{1}{|G_b - G_u|} \sqrt{\frac{2}{Le_G} \int_{G_u}^{G_b} \left(\frac{\sigma}{\sigma_b} \right)^2 \frac{\lambda}{c_p} w_G dG}. \quad (32)$$

Thus, $\mathcal{F}(\boldsymbol{\psi}_b; G_u)$ is the mass burning rate of a stretchless flame with infinitely thin reaction layer. Note, that we explicitly emphasize the dependence of the mass burning rate on the state $\boldsymbol{\psi}_b^0 = (h_b^0, Z_{1,b}^0, \dots, Z_{N_e-1,b}^0)$ in the burnt gas. This follows from the observation that the factor $\frac{\lambda}{c_p} w_G$ in equation (32) is a function of the (stretchless) combustion variables Y_i and T . However, the integral in (32) effectively only runs over the thin reaction layer, where $w_G \neq 0$ and the mixture approaches the equilibrium state. From a time scale analysis of a few combustion systems, it is clear that the reaction path in composition space is embedded in a low-dimensional manifold, which can be described by a small number of progress variables [7, 15]. Near the equilibrium point in composition space, all reaction paths are mostly attracted to a one-dimensional subspace, independent of the initial composition. This means that in this case all combustion variables can be written as a function of the variable G , because $\nabla G \neq \mathbf{0}$. The integral over G in (32) is thus independent of the initial composition and depends only on the equilibrium state, which is completely determined by the pressure p_{amb} , the enthalpy h_b^0 and the element mass fractions $Z_{b,j}^0$. Finally, the dependence of m_b^0 on G_u^0 comes from the factor $1/|G_b - G_u|$ in front of the square root in (32).

Next, we consider stretched flames, i.e. $K(s) \neq 0$. Integration of the flamelet equations (24) for mass, enthalpy and element mass fractions now gives the following integral balances:

$$\begin{aligned} (\sigma m)_b - (\sigma m)_u &= - \int_{s_u}^{s_b} \sigma \rho K \, ds, \\ (\sigma m)_b (h_b - h_u) &= - \int_{s_u}^{s_b} \sigma \rho K (h - h_u) \, ds, \\ (\sigma m)_b (Z_{j,b} - Z_{j,u}) &= - \int_{s_u}^{s_b} \sigma \rho K (Z_j - Z_{j,u}) \, ds, \quad j = 1, \dots, N_e - 1. \end{aligned} \quad (33)$$

As we have seen that the mass burning rate m_b depends on the enthalpy and the element composition in the burnt gas, we have to compute the differences $h_b - h_u$ and $Z_{j,b} - Z_{j,u}$. Substitution of relation (12) into the second equation of (33) and definition equation (14) of Z_j into the third equation gives:

$$\begin{aligned} (\sigma m)_b (h_b - h_u) &= -(h_{T,b} - h_{T,u}) \int_{s_u}^{s_b} \sigma \rho K \tilde{h}_T \, ds - \sum_{i=1}^{N_s-1} h_i^0 (Y_{i,b} - Y_{i,u}) \int_{s_u}^{s_b} \sigma \rho K \tilde{Y}_i \, ds, \\ (\sigma m)_b (Z_{j,b} - Z_{j,u}) &= - \sum_{i=1}^{N_s-1} w_{j,i} (Y_{i,b} - Y_{i,u}) \int_{s_u}^{s_b} \sigma \rho K \tilde{Y}_i \, ds, \quad j = 1, \dots, N_e - 1, \end{aligned} \quad (34)$$

where $\tilde{f}(s) = (f(s) - f_u)/(f_b - f_u)$ is the normalized variable corresponding with f ($f = Y_i, h_T$). The first equation in (34) relates the enthalpy difference $h_b - h_u$ to the stretch term $\sigma \rho K$ through out the flame, weighted with the normalized combustion variables \tilde{h}_T and \tilde{Y}_i . A

similar conclusion holds for the differences $Z_{j,b} - Z_{j,u}$ of the element mass fractions.

We now proceed with the computation of the mass burning rate m_b , which can be computed again from equation (27), with $\mathcal{C}(s)$ replaced by

$$\mathcal{C}(s) = \frac{\partial}{\partial s}(\sigma m G) + \sigma \rho K G, \quad (35)$$

while $\mathcal{D}(s)$ and $\mathcal{B}(s)$ are still defined in (28) (with the superscripts '0' omitted). Analogously to the computation of m_b^0 we find

$$m_b + \frac{1}{\sigma_b} \int_{s_u}^{s_b} \sigma \rho K \tilde{G} ds \approx \mathcal{F}(\boldsymbol{\psi}_b; G_u). \quad (36)$$

The term in the right-hand side is the approximation of the mass burning rate of a stretchless flame, with enthalpy and element composition given by h_b and $Z_{j,b}$ instead of h_b^0 and $Z_{j,b}^0$.

These equations can be further elaborated if we introduce the Karlovitz numbers. The Karlovitz number Ka_f associated with the variable f ($f = Y_i, h_T$) is defined by

$$Ka_f = Le_f \int_{s_u}^{s_b} \sigma \rho K \tilde{f} ds / \sigma_b \mathcal{F}(\boldsymbol{\psi}_b; G_u), \quad (37)$$

i.e. Ka_f is the mass flow rate $\sigma \rho K$ along the flame surfaces weighted with $Le_f \tilde{f}$ and integrated over the flame, scaled with the mass flow rate $\sigma_b \mathcal{F}(\boldsymbol{\psi}_b; G_u)$. The variable \tilde{f} in (37) has to be computed (numerically) from the corresponding flamelet equation in (24). Introducing definition equation (37) of the Karlovitz numbers in the balances for enthalpy and element mass fractions in (34), and subsequently eliminating $h_{T,b} - h_{T,u}$ using equation (12), we obtain the following relations:

$$\begin{aligned} (m_b + Ka_{h_T} \mathcal{F}(\boldsymbol{\psi}_b; G_u))(h_b - h_u) &= \mathcal{F}(\boldsymbol{\psi}_b; G_u) \sum_{i=1}^{N_s-1} \left(Ka_{h_T} - \frac{Ka_i}{Le_i} \right) h_i^0 (Y_{i,b} - Y_{i,u}), \\ (m_b + Ka_{h_T} \mathcal{F}(\boldsymbol{\psi}_b; G_u))(Z_{j,b} - Z_{j,u}) &= \mathcal{F}(\boldsymbol{\psi}_b; G_u) \sum_{i=1}^{N_s-1} \left(Ka_{h_T} - \frac{Ka_i}{Le_i} \right) w_{j,i} (Y_{i,b} - Y_{i,u}), \\ & j = 1, \dots, N_e - 1, \end{aligned} \quad (38)$$

where we use the notation Ka_i instead of Ka_{Y_i} . Likewise, equation (36) for the mass burning rate simply reduces to

$$m_b \approx \left(1 - \frac{Ka_G}{Le_G} \right) \mathcal{F}(\boldsymbol{\psi}_b; G_u). \quad (39)$$

With the obvious choice $G = h_T$, suggested by the expression for m_b in (39), the equations in (38) reduce to the approximations:

$$h_b - h_u \approx \sum_{i=1}^{N_s-1} \left(K a_{h_T} - \frac{K a_i}{L e_i} \right) h_i^0 (Y_{i,b} - Y_{i,u}),$$

$$Z_{j,b} - Z_{j,u} \approx \sum_{i=1}^{N_s-1} \left(K a_{h_T} - \frac{K a_i}{L e_i} \right) w_{j,i} (Y_{i,b} - Y_{i,u}), \quad j = 1, \dots, N_e - 1. \quad (40)$$

The relations in (40) describe the effect of flame stretch and preferential diffusion on the enthalpy and element composition in the burnt gas. Summarizing, we have to solve equations (39) and (40) for the computation of the mass burning rate, the enthalpy and the element mass fractions in the burnt gas mixture, with the Karlovitz numbers defined in (37). Furthermore, computation of the Karlovitz numbers requires the numerical solution of the flamelet equations (24). A numerical validation of this model is presented in [10]. In this paper, we pursue an analytical approach. We approximate the Karlovitz numbers using the stretchless solution of the flamelet equations and subsequently we can explicitly compute the mass burning rate from the set (39)-(40) for the special case of lean methane/air flames. The results we obtain this way are compared with existing theory, and are presented in the next section.

5 Mass burning rate of lean methane/air flames

In this section we apply the results of Section 4 to lean methane/air flames. We first calculate the stretchless mass burning rate and subsequently study the influence of flame stretch. Herewith, we make the following assumptions:

1. The flames we consider are locally flat, i.e. $\sigma(s) = 1$.
2. The specific heats of all species are equal and constant, i.e. $c_{p,i} = c_p = \text{const}$ for all i .
3. The thermal conductivity λ is constant.
4. The stretch term ρK is constant, i.e. $\rho K = (\rho K)_b$.
5. The flames are weakly stretched, so that all Karlovitz numbers are small.

As a consequence of the second assumption, the third term in the left-hand side of the flamelet equation for h_T in (24) vanishes. Moreover, it is clear that $h_T = c_p(T - T_0)$ and that $K a_{h_T} = K a_T$, and for this reason we choose $G = T$, instead of $G = h_T$, in this section.

Consider a lean methane/air flame described by the single-step irreversible reaction $CH_4 + aO_2 \rightarrow CO_2 + 2H_2O + (a-2)O_2$, ($a > 2$). The mass burning rate m_b^0 follows from equations (31) and (32) with $w_G = w_{h_T}/c_p$. For the source term w_{h_T} we adopt the model

$$w_{h_T} = \Delta H A \rho^{\alpha+\beta} Y_{CH_4}^\alpha Y_{O_2}^\beta \exp(-E_a/RT), \quad (41)$$

proposed by de Lange et al. [12]. The reaction rate parameters α , β , E_a and A are fitted to experimental data of the mass burning rate. Substitution of (41) into (32) gives $m_b^0 \approx \mathcal{F}(\boldsymbol{\psi}_b^0; T_u^0)$ with

$$\mathcal{F}(\boldsymbol{\psi}_b; T_u) = \left(2A\Gamma(\alpha + 1) \frac{\lambda}{c_p} L e_{CH_4}^\alpha\right)^{\frac{1}{2}} Y_{O_2,b}^{\beta/2} \rho_b^{(\alpha+\beta)/2} \times \frac{T_b}{T_b - T_u} \left(\frac{c_p T_b}{\Delta H}\right)^{(\alpha-1)/2} \left(\frac{RT_b}{E_a}\right)^{(\alpha+1)/2} \exp(-E_a/2RT_b). \quad (42)$$

In the derivation of (42), we applied the Frank-Kamanetskii transformation [25] and used the relations $\rho = \rho_b$, $Y_{CH_4} = L e_{CH_4} c_p (T_b - T) / \Delta H$ and $Y_{O_2} = Y_{O_2,b}$ which approximately hold near the reaction layer.

For stretched flames we have to compute the mass burning rate m_b from equations (39) and (40), with the Karlovitz numbers defined in (37). Since the flames are weakly stretched, we replace $\tilde{f}(s)$ ($f = T, Y_i$) in (37) by its stretchless counterpart $\tilde{f}^0(s)$, which can be computed from the corresponding flamelet equation. Furthermore, we neglect the source terms for T and Y_i , which means that we effectively compute these variables in the preheat zone. This way, we obtain the approximations

$$Ka_T, Ka_i \approx Ka = \frac{(\rho K)_b}{(m_b^0)^2} \frac{\lambda}{c_p}, \quad i = 1, \dots, N_s - 1. \quad (43)$$

Comparing the mass burning rate $m_b(\boldsymbol{\psi}_b; T_u)$ of a stretched (stagnation) flame with the mass burning rate $m_b^0(\boldsymbol{\psi}_b^0; T_u)$ of a stretchless flame, with both flames having the same temperature in the unburnt gas mixture, i.e. $T_u = T_u^0$, and taking into account that $h_b^0 = h_u$ and $Z_{j,b}^0 = Z_{j,u}$ ($j = 1, \dots, N_e - 1$), we can derive the first order approximation

$$\frac{m_b(\boldsymbol{\psi}_b; T_u)}{m_b^0(\boldsymbol{\psi}_b^0; T_u)} \approx 1 - Ka + \Delta h \frac{\partial}{\partial h_b} \ln \mathcal{F}(\boldsymbol{\psi}_b^0; T_u) + \sum_{j=1}^{N_e-1} \Delta Z_j \frac{\partial}{\partial Z_{j,b}} \ln \mathcal{F}(\boldsymbol{\psi}_b^0; T_u) + \mathcal{O}(Ka^2), \quad (44)$$

with the differences Δh and ΔZ_j given by

$$\Delta h = h_b - h_u \approx Ka \sum_{i=1}^{N_s-1} \left(1 - \frac{1}{Le_i}\right) h_i^0 (Y_{i,b} - Y_{i,u}) + \mathcal{O}(Ka^2),$$

$$\Delta Z_j = Z_{j,b} - Z_{j,u} \approx Ka \sum_{i=1}^{N_s-1} \left(1 - \frac{1}{Le_i}\right) w_{j,i} (Y_{i,b} - Y_{i,u}) + \mathcal{O}(Ka^2), \quad j = 1, \dots, N_e - 1. \quad (45)$$

Substituting the approximations of Δh and ΔZ_j into equation (44), we obtain

$$\frac{m_b(\boldsymbol{\psi}_b; T_u)}{m_b^0(\boldsymbol{\psi}_b^0; T_u)} \approx 1 - \mathcal{M}Ka + \mathcal{O}(Ka^2), \quad (46)$$

where the Markstein number \mathcal{M} is given by

$$\mathcal{M} = 1 + \sum_{i=1}^{N_s-1} \left(\frac{1}{Le_i} - 1 \right) (Y_{i,b} - Y_{i,u}) \left(h_i^0 \frac{\partial}{\partial h_b} + \sum_{j=1}^{N_e-1} \frac{\partial}{\partial Z_{j,b}} \right) \ln \mathcal{F}(\boldsymbol{\psi}_b^0; T_u). \quad (47)$$

Evaluation of equation (47) requires the computation of the partial derivatives of $\ln \mathcal{F}(\boldsymbol{\psi}_b; T_u)$ with respect to h_b and $Z_{j,b}$. However, in equation (42) the mass burning rate $\mathcal{F}(\boldsymbol{\psi}_b; T_u)$ is expressed in terms of T_b and $Y_{i,b}$. The partial derivatives in (47) can only be computed if there exists a one-to-one relation between the set of state variables $\boldsymbol{\psi}_b = (h_b, Z_{j,b})$ on the one hand and the combustion variables $\boldsymbol{\phi}_b = (T_b, Y_{i,b})$ on the other hand. The equilibrium relations have to be used to define this relation. Since $Y_{CH_4,b} = 0$ and N_2 is inert, the following relations hold for the equilibrium state of methane/air flames

$$\begin{aligned} h_b &= c_p(T_b - T_0) + h_{O_2}^0 Y_{O_2,b} + h_{CO_2}^0 Y_{CO_2,b} + h_{H_2O}^0 Y_{H_2O,b}, \\ Z_{C,b} &= w_{C,CO_2} Y_{CO_2,b}, \\ Z_{H,b} &= w_{H,H_2O} Y_{H_2O,b}, \\ Z_{O,b} &= w_{O,O_2} Y_{O_2,b} + w_{O,CO_2} Y_{CO_2,b} + w_{O,H_2O} Y_{H_2O,b}. \end{aligned} \quad (48)$$

Using the chain rule and the implicit function theorem, the derivatives in (47) can be expressed in terms of the derivatives $\frac{\partial}{\partial T_b} \ln \mathcal{F}(\boldsymbol{\psi}_b; T_u)$ and $\frac{\partial}{\partial Y_{i,b}} \ln \mathcal{F}(\boldsymbol{\psi}_b; T_u)$. The only relevant partial derivatives in our case are

$$\begin{aligned} \frac{\partial}{\partial T_b} \ln \mathcal{F}(\boldsymbol{\psi}_b; T_u) &= \frac{1}{2(T_b - T_u)} \left(\alpha - \beta - (\alpha - \beta + 2) \frac{T_u}{T_b} + Ze \right), \\ \frac{\partial}{\partial Y_{O_2,b}} \ln \mathcal{F}(\boldsymbol{\psi}_b; T_u) &= \frac{\beta}{2Y_{O_2,b}}, \end{aligned} \quad (49)$$

with $Ze = E_a(T_b - T_u)/(RT_b^2)$ the Zeldovich number and consequently equation (47) for the Markstein number reduces to

$$\begin{aligned} \mathcal{M} &= 1 - \left(\frac{1}{Le_{CH_4}} - 1 \right) \frac{\Delta H}{c_p} \frac{Y_{CH_4,u}}{M_{CH_4}} \frac{\partial}{\partial T_b} \ln \mathcal{F}(\boldsymbol{\psi}_b^0; T_u) + \\ &\left(\left(\frac{1}{Le_{CH_4}} - 1 \right) \frac{2M_{O_2}}{M_{CH_4}} Y_{CH_4,u} + \left(\frac{1}{Le_{O_2}} - 1 \right) (Y_{O_2,b} - Y_{O_2,u}) \right) \frac{\partial}{\partial Y_{O_2,b}} \ln \mathcal{F}(\boldsymbol{\psi}_b^0; T_u), \end{aligned} \quad (50)$$

where $\Delta H = M_{CH_4} h_{CH_4}^0 + 2M_{O_2} h_{O_2}^0 - M_{CO_2} h_{CO_2}^0 - 2M_{H_2O} h_{H_2O}^0$ is the heat of combustion. Finally, if we substitute the partial derivatives in (49) into equation (50) and if we replace the combustion variables by their stretchless counterparts, using the relations $h_b^0 = h_u$ and $Z_{j,b}^0 = Z_{j,u}$ ($j = 1, \dots, N_e - 1$), we find

$$\mathcal{M} = 1 - \left(\frac{1}{Le_{CH_4}} - 1 \right) \frac{1}{2} (\alpha - \beta - (\alpha - \beta + 2) \frac{T_u}{T_b^0} + Ze^0) - \left(\frac{1}{Le_{CH_4}} - \frac{1}{Le_{O_2}} \right) (Y_{O_2,u}^0 - Y_{O_2,b}^0) \frac{\beta}{2Y_{O_2,b}^0}. \quad (51)$$

From this equation for the Markstein number and equation (46) we see that variations in the mass burning rate are caused by flame stretch, possibly in combination with preferential diffusion. To be more specific, the second term on the right-hand side of equation (51) gives the effect of the flame temperature on the variation of the mass burning rate, and is due to the difference between the diffusivity of the lean species (CH_4) and the conductivity of the gas mixture. Furthermore, the Lewis number of CH_4 is the only one which appears in this term. This observation is clear from a physical point of view, because temperature variation by preferential diffusion is obviously independent of the Lewis number of O_2 in a bulk of air and also of the Lewis numbers of the product species CO_2 and H_2O in a lean irreversible reaction, in which the products play no role in the propagation speed of the flame. The third term on the right-hand side of equation (51) is related to local stoichiometry changes by differential diffusion, and plays a role when more than one reactant species is considered. The factor $1/Le_{CH_4} - 1/Le_{O_2}$ in this term is related to the difference in diffusivities of the reactant species.

Although a lot of data concerning the mass burning rate are available in literature, a comparison of equations (51) and (46) with existing expressions for the mass burning rate is difficult, because our approach differs from what most other authors use. Without trying to be complete, we just mention a few results. Clavin et al. [5] derived an expression for the Markstein number in case of an irreversible one-step reaction, based on large activation energy asymptotics. They found an expression, describing the effect of thermal expansion on the Markstein number which is analogous to the first two terms in (51). In [4], Chung et al. derived an expression for the mass burning rate, using an integral analysis of the conservation laws of laminar flames, and they found an expression comparable to (51). Our analysis is also very similar to the 1D stream tube model of Echehki, presented in [6]. He derived a general expression for the mass burning rate, comparable with (32), from quasi-one-dimensional conservation laws, which hold for a small stream tube in the flame. However, he does not present the analogon of equation (51), which holds for the special case of a lean methane/air flame.

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