Detailed analysis of the mass burning rate of stretched flames including preferential diffusion effects

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Abstract

This study focuses on the effects of flame stretch on the mass burning rate, for flames with nonunit Lewis numbers. The extended flame stretch model of de Goey and ten Thije Boonkkamp [Combust. Flame 119 (1999) 253–271], which was derived for multiple-species transport and chemistry, is used as a starting point. This model is adjusted to be able to predict the mass burning rate at the inner reaction layer of the flame. The accuracy of the theory is analyzed step by step using numerical results incorporating detailed chemistry and multispecies transport models. The adjusted model proves to be an accurate predictor of the mass burning rate. Furthermore, it is shown that not only the lean species Lewis number plays a role but also a number of other Lewis numbers have a significant influence on preferential diffusion. Preferential diffusion for methane/air mixtures is more difficult to predict accurately than for ethane/air and propane/air mixtures. Different contributions to the total preferential diffusion effect in methane/air mixtures partly cancel, which corresponds to an effective Lewis number being close to one. For fuels with Lewis numbers further away from one, no cancellation takes place and the accuracy of the predicted mass burning rate increases. Results show that the extended theory forms the basis for the first quantitative model to predict mass burning rates of premixed, laminar, stretched flames with nonunit Lewis numbers.

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Keywords: Laminar premixed flames; Mass burning rate; Flame stretch; Preferential diffusion

1. Introduction

It is well known that flame stretch is a very important quantity in combustion science. Karlovitz et al. [1] and Markstein [2] were the first to recognize the effect of flame stretch on premixed flame structures and dynamics. Flame stretch has been studied by many researchers since, such as Buckmaster [3,4], Matalon [5], Matalon and Matkowsky [6], and Chung and Law [7]. A rigorous mathematical analysis of stretched flames, using matched asymptotic expansions, was introduced by Matalon and Matkowsky [6]. An integral analysis of flame stretch was presented by Chung and Law [7]. It must be stressed that the global
physical influence of flame stretch on flame dynamics is known at the moment, but one must realize that most of the studies performed so far were related to flames described by global kinetics and simple transport models. It is not known at the moment to what extent current models are able to describe or predict the influence of stretch on real flames with detailed chemistry and multiple-species transport models. For example, in most existing theories, the Lewis number of the lean species in the unburnt mixture is the only Lewis number which plays a role, whereas the influence of other species including radicals and intermediates is not included. Chung et al. [8] quantified the effect of stretch and preferential diffusion using a local equilibrium temperature for hydrogen–air premixed flames. Recently de Goey and ten Thije Boonkkamp [9] derived a premixed flamelet model from the full set of 3D time-dependent conservation equations. From these flamelet equations, a general expression for the mass burning rate has been derived from first principles extending the integral analysis, introduced by Chung and Law [7]. It is expected that this model can be used as a basis for a quantitative model describing the influence of flame stretch and preferential diffusion on premixed flame dynamics. In previous studies it has been shown already that this model accurately describes the influence of flame stretch on premixed laminar flames [10], partially premixed triple flames [11], and even in case of highly stretched turbulent flames [12] for unit Lewis numbers. The application of the theory to detailed chemistry with nonunit Lewis numbers has not been studied in detail so far. The focus of this paper is on this aspect for the case of weakly stretched planar flames. In the future, the application to strong stretch will be analyzed.

In the next section, the basic model of de Goey and ten Thije Boonkkamp will briefly be reviewed and the model will be adjusted to describe the influence of flame stretch on the mass burning rate at the inner layer rather than at the burnt side of the flame. It is known that preferential diffusion in combination with flame stretch introduces a change in the element composition and enthalpy at the inner reaction layer. As the burning velocity is sensitive to these changes, this influence will be studied in the subsequent section. This part of the study is based on numerical results of lean adiabatic methane/air, ethane/air, and propane/air flames without flame stretch. It is observed that the coefficients that describe the sensitivity of the mass burning rate to changes in enthalpy and elemental composition are almost independent of the fuel and stoichiometry used. Models for the change in enthalpy and elemental composition are presented in the subsequent section. In the same section the accuracy of the expressions describing the change in enthalpy and the element mass fraction due to preferential diffusion and flame stretch will be analyzed and compared with direct numerical results. These changes in enthalpy and element composition at the inner layer, combined with the sensitivities of the mass burning rate on such changes, finally induce a change in the stretched mass burning rate, which is studied in the last section. Deviations between theory and numerical simulations will be identified as well. The paper ends with a discussion on the species that dominate the behavior and conclusions from the analysis.

The present study is a continuation of the work reported in a previous paper by de Goey and ten Thije Boonkkamp [9]. Compared to that paper several new elements are presented here. First, the theory introduced in [9] is adapted in order to describe the effect of flame stretch and preferential diffusion on the mass burning rate at the inner layer instead of at the burnt side of the flame. This position is found to be more relevant to the propagation speed of the flame as explained in the next section. Second, we study a linearized version of the original expression for the mass burning rate for the case of weak stretch. This linearized expression was already proposed in [9] but it was not compared with numerical simulations. Due to this linearization, sensitivity coefficients appear, which describe the effect of enthalpy and element mass fraction changes on the mass burning rate. In the nonlinearized expression this effect was taken into account by a direct numerical simulation of an unstretched flame with changed enthalpy and element composition. Finally, in [9] the theory was validated against numerical simulations of lean methane–air flames, in which the preferential diffusion effect appeared to be very small. Therefore, this study is also broadened in the sense that two additional fuels (ethane and propane) are investigated, which are known to be more sensitive to preferential diffusion effects.

2. Mass burning rate of stretched flames

Based on the full set of 3D unsteady conservation equations for deflagration processes, de Goey and ten Thije Boonkkamp derived a set of flamelet equations for premixed flames [9]. The basic ideas will be summarized in this section and we will derive an adjusted model. The flame is defined in terms of iso-surfaces of a progress variable \( \chi \) and a local orthogonal coordinate transformation is used to define a coordinate \( s \) perpendicular to these surfaces. We can rewrite mass conservation as follows,

\[
\frac{\partial}{\partial s} (\sigma m) = -\sigma \rho K, \tag{1}
\]
where \( m = \rho s_L \) is the mass burning rate, \( s_L \) is the burning velocity, \( \rho \) is the mass density, and \( \sigma \) is an area function. The stretch term on the right-hand side corresponds to the leaking of mass in the two directions perpendicular to coordinate \( s \) (i.e., along the flame surfaces). Conservation of chemical species \( i \), represented by species mass fraction \( Y_i \), and conservation of enthalpy, \( h \), can be written as

\[
\frac{\partial}{\partial s}(\sigma m Y_i) - \frac{1}{Le_i} \frac{\partial}{\partial s}\left( \sigma \frac{\lambda}{c_p} \frac{\partial Y_i}{\partial s} \right) - \sigma \dot{Y}_i = -\sigma \rho K Y_i
\]

\((i = 1, \ldots, N_s - 1)\) \hspace{1cm} (2)

and

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

\[(3)\]

In these equations we can clearly identify convection, which is the first term on the left-hand sides, and diffusion, which is the second term in Eq. (2) and the second and third term in Eq. (3). Furthermore there is a source term \( \dot{Y}_i \) of species \( i \), because species can be created and consumed by chemical reactions. Again the stretch terms on the right-hand sides correspond to transport of chemical species and enthalpy in the two directions perpendicular to \( s \). \( Le_i \) corresponds to the constant Lewis number of species \( i \), \( \lambda \) is the thermal conductivity, \( c_p \) is the specific heat at constant pressure, and \( h_i \) is the specific enthalpy of species \( i \). The element mass fraction \( Z_j \) of element \( j \) is defined by

\[
Z_j = \sum_{i=1}^{N_s} W_{ji} Y_i \quad (j = 1, \ldots, N_e),
\]

\[(4)\]

with \( W_{ji} \) the mass of element \( j \) per mass of species \( i \). By taking a suitable linear combination of Eq. (2), we obtain

\[
\frac{\partial}{\partial s}(\sigma m Z_j) - \frac{\partial}{\partial s}\left( \sigma \frac{\lambda}{c_p} \frac{\partial Z_j}{\partial s} \right)
\]

\[
- \frac{\partial}{\partial s}\left( \sigma \frac{\lambda}{c_p} \sum_{i=1}^{N_s} \left( \frac{1}{Le_i} - 1 \right) W_{ji} \frac{\partial Y_i}{\partial s} \right) = -\sigma \rho K Z_j.
\]

\[(5)\]

In Eq. (5) the only source term is presented on the right-hand side. Elements cannot be created or consumed by chemical reactions, but there is leaking of element \( j \) along the flame surfaces. The flame stretch rate \( K \) is defined as the relative rate of change of mass \( M \) in a small control volume inside the flame structure,

\[
K = \frac{1}{M} \frac{dM}{dt},
\]

\[(6)\]
as derived in [13]. In general we can say that transport terms along the flame surfaces are gathered in the terms proportional to \( \rho K \). Terms that are due to non-steady effects in the flame-adapted coordinate system and terms arising because local iso-surfaces of the variables \( Y_i, h, \) and \( Z_j \) do not necessarily coincide are neglected. This is valid when the flame structure is thin compared to external distortions. This was investigated by de Goey and ten Thije Boonkkamp [9]. They computed the magnitude of these terms and they appeared to be very small.

In this study we will investigate planar flames, which means that \( \sigma \) is equal to one (\( \sigma = 1 \)) and we will focus on weak stretch. Curved flames and strong stretch will be studied in the future. An expression for the mass burning rate at the burnt side, indicated by subscript \( b \), of a weakly stretched premixed flame has been derived from the flamelet equations [9]

\[
m_b \approx -\frac{1}{Le} + \Delta h \frac{\partial}{\partial \ln \left( m_0^b \right)} \left( \ln \left( m_0^b \right) \right) + \sum_{j=1}^{N_e} \frac{\partial}{\partial \ln Z_j^b} \left( \ln \left( m_0^b \right) \right) + \text{h.o.t.,}
\]

\[(7)\]

in which the Karlovitz integral, \( K_{\text{y}} \), is defined as

\[
K_{\text{y}} = \frac{\text{Le}}{m_0^b} \int_{s_b}^{s_0} \rho K \left( \lambda - \lambda_u \right) ds,
\]

\[(8)\]

where \( \lambda \) is the progress variable. The subscript \( u \) indicates the unburnt side of the flame. The enthalpy and element mass fraction changes are defined as

\[
\Delta h = h(s) - h^0(s),
\]

\[(9)\]

\[
\Delta Z_j(s) = Z_j(s) - Z_j^0(s).
\]

\[(10)\]

In Eq. (7) the changes at the burnt side of the flame are used (\( s = s_b \)). To arrive at expression (7) it has been assumed that the chemical source term is only important in a thin reaction layer within the total flame structure. This assumption can be satisfied by choosing a proper progress variable. In this study we used the fuel mass fraction as progress variable following [9], because it is rapidly consumed in a thin layer. Since it is known that the reacting paths in composition space are attracted to a one-dimensional manifold close to the equilibrium point [14], the mass burning rate is assumed to depend only on the enthalpy and element composition at the burnt side of the flame (\( m_0^b \approx \mathcal{F}(\lambda, h^0, Z_{1,b}^0, \ldots, Z_{N_e,b}^0) \)) [9].

In the present study, the above model will be adjusted to an expression for the mass burning rate at the inner layer position, indicated by subscript \( il \). The inner layer is physically more relevant than the burnt side of the flame, where chemical equilibrium is
reached. It must be noted that equilibrium is reached more than 10 flame thicknesses away from the inner reaction layer. Therefore, in this study the burnt side indicates a position relatively far downstream of the flame. The inner layer is the region where most chemical activity takes place. So, the mass burning rate at the inner layer gives a better representation of the propulsion speed of the flame than at any other position. Moreover, Groot and de Goey [15] have shown that the unstretched mass burning rate \( m_0 \) is independent of curvature effects, only when it is evaluated at the inner layer. Therefore, \( m_0 \) does depend on curvature effects, while \( m_0^0 \) does not. As a consequence, we expect that the enthalpy and element composition close to the inner layer determine the burning velocity rather than the composition at the burnt side of the flame: \( m_0^0 \approx F(Y_{U}, h_{i}^0, \sigma_{1,il^0}, \ldots, Z_{N_e,il^0}). \)

The model by de Goey and ten Thije Boonkkamp [9] is derived from the expression

\[
m_b = m_b^0(Y_u, Z_b, h_b) - \int_{s_u}^{s_b} \sigma K \left( \frac{\gamma - \gamma _u}{\gamma_b - \gamma_u} \right) ds, \tag{11}
\]

which we will use to derive the adjusted model as well. The mass burning rate of unstretched flames is constant through the flame which means that \( m_{iil} = m_0^0 \). Together with the integrated form of Eq. (1),

\[
m_{iil} = m_b + \int_{s_{iil}}^{s_b} \rho K ds, \tag{12}
\]

we can combine this with Eq. (11) and use a linear Taylor expansion to derive

\[
m_{iil} = \frac{1}{m_0^0} - \frac{K \gamma_{il}}{L \gamma} + \Delta h_{iil} \frac{\partial}{\partial h_{iil}^0} (\ln m_{iil}^0) \]

\[
+ \sum_{j=1}^{N_e-1} \Delta Z_{j,il} \frac{\partial}{\partial Z_{j,il}^0} (\ln m_{iil}^0) + h.o.t. \tag{13}
\]

The adjusted Karlovitz integral, \( K_{\gamma_{il}} \), is now defined as

\[
K_{\gamma_{il}} = \frac{L \gamma}{m_{iil}^0} \int_{s_u}^{s_b} \rho K \left[ \left( \frac{\gamma - \gamma _u}{\gamma_b - \gamma_u} \right) - H(s - s_{iil}) \right] ds, \tag{14}
\]

where \( H(s - s_{iil}) \) is the Heaviside function, equal to zero when \( s < s_{iil} \) and equal to 1 when \( s > s_{iil} \). It can be shown that the model presented in this paper yields the same expression for the Markstein number as found in [9]. Under the assumptions of one-step chemistry, a single Lewis number, exponential temperature and species profiles in the flame, and a thin reaction layer, de Goey and ten Thije Boonkkamp [9] found expressions for the Markstein number that were also found by Joulin and Clavin [16] and Clavin [17]. Equation (13) is the heart of the adjusted model we want to analyze in the next few sections. This will be done in three steps: First, the sensitivity of the unstretched mass burning rate \( m_0^0 \) to changes in enthalpy and element mass fractions will be analyzed. Second, the changes in enthalpy \( \Delta h \) and element mass fractions \( \Delta Z_{j} \) due to weak stretch effects will be analyzed. Third, the resulting accuracy of the prediction of the mass burning rate will be examined.

3. Sensitivity of the unstretched mass burning rate \( m_{iil}^0 \) to changes in \( h \) and \( Z_j \)

The burning velocity of an unstretched adiabatic planar flame changes if the enthalpy and/or element composition is adapted. For convenience the preferential diffusion terms in Eq. (13) will be referred to as \( P_{\gamma} \) and \( P_{\gamma} \).

\[
P_h = \Delta h_{iil} \frac{\partial}{\partial h_{iil}^0} (\ln m_{iil}^0) = c_h \Delta h_{iil} \tag{15}
\]

and

\[
P_j = \Delta Z_{j,il} \frac{\partial}{\partial Z_{j,il}^0} (\ln m_{iil}^0) = c_j \Delta Z_{j,il} \tag{16}
\]

for element index \( j = C, O, H \). Note that there are only \( N_e - 1 \) independent changes \( \Delta Z_j \). The change of the last element mass fraction—in our case that of nitrogen \( \Delta Z_N \)—is given by

\[
\Delta Z_N = - \sum_{j=1}^{N_e-1} \Delta Z_j. \tag{17}
\]

A numerical approach is used to determine the values of the coefficients \( c_h \) and \( c_j \) that appear in Eqs. (15) and (16). The change in mass burning rate depends on changes in enthalpy and element composition. However, the mass burning rate is independent of the cause of \( \Delta h_{iil}^0 \) and \( \Delta Z_{j,il}^0 \); it only depends on the magnitude of these parameters. To determine the sensitivity coefficients \( c_h \) and \( c_j \), we used unstretched flames and changed several input parameters (temperature and composition of the unburnt mixture) to generate different values for \( \Delta h_{iil}^0 \) and \( \Delta Z_{j,il}^0 \). It must be stressed that \( \Delta h_{iil}^0 \) and \( \Delta Z_{j,il}^0 \) themselves are not input parameters of a flame simulation, but a consequence of a change in input parameters. For this reason, classical sensitivity analysis tools cannot be used. A set of adiabatic unstretched premixed flat flame solutions of Eqs. (1–3) is generated using a
Table 1
Sensitivity coefficients

<table>
<thead>
<tr>
<th>Variable</th>
<th>Methane</th>
<th>Ethane</th>
<th>Propane</th>
</tr>
</thead>
<tbody>
<tr>
<td>c_h (g/kJ)</td>
<td>1.5</td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>c_C (-)</td>
<td>36.8</td>
<td>23.3</td>
<td>34.6</td>
</tr>
<tr>
<td>c_O (-)</td>
<td>3.1</td>
<td>6.9</td>
<td>-2.1</td>
</tr>
<tr>
<td>c_H (-)</td>
<td>104.0</td>
<td>154.3</td>
<td>142.2</td>
</tr>
</tbody>
</table>

flamelet code [18] and the changes in enthalpy (Δh^0_j) and element mass fraction (ΔZ^0_j) as well as the corresponding mass burning rate (m^0_j) are recorded. The resulting (overdetermined) set of linear equations is solved to determine the sensitivity coefficients. In this study, methane, ethane, and propane flames are used, having a stoichiometric ratio of φ = 0.8 as reference case. Also a methane flame, having a stoichiometric ratio of φ = 0.9, is studied. Constant Lewis numbers are used and they are determined following the procedure described in [19]. The reaction mechanism that is used consists of 39 species and 173 reversible reactions [20]. A total of 80 computations per reference case is performed. The sensitivity coefficients are presented in Table 1.

Taking a subset of the 80 solutions generated gives approximately the same values for the sensitivity coefficients. This confirms the validity of the assumption that the mass burning rate primarily depends on the enthalpy and element mass fractions at the inner layer and does not depend on the cause of Δh^0_j and/or ΔZ^0_j,i,l. Furthermore it can be seen that the coefficient values for enthalpy and the different elements are almost independent of the fuel and stoichiometric ratio. This might imply that the fuel breaks up very fast and then follows a universal path, which is approximately the same for different hydrocarbon fuels. This corresponds to low-dimensional manifold theories [14]. This is an interesting result, which requires further study.

4. The influence of stretch on the enthalpy and element mass fractions

The accuracy of the model for the stretched mass burning rate depends on the accuracy of the prediction of the enthalpy and element mass fraction changes. In [9], expressions have been presented for Δh^0_j and ΔZ^0_j,b derived by integrating the conservation equations of enthalpy and element mass fractions over the flame up to the burnt side. Here we present a general expression and we will compare the results for the burnt side and the inner layer. The elemental mass fractions Z^0_j(s) at position s in an unstretched flame are given by

\[ Z^0_j(s) - Z_{j,u} = \frac{D_j^0}{m^0_j} \]  

and for a stretched flame,

\[ Z_j(s) - Z_{j,u} = -\frac{1}{m} \int_{s_u}^{s} \rho K(Z_j - Z_{j,u}) \, ds + \frac{D_j}{m}. \]  

The expression for D_j^0 is similar to the previous expression, but using values from an unstretched flame instead of a stretched one. Subtracting Eq. (18) from Eq. (19) leads to

\[ \Delta Z_j(s) = Z_j(s) - Z^0_j(s) = -\frac{1}{m} \int_{s_u}^{s} \rho K(Z_j - Z_{j,u}) \, ds + \frac{D_j}{m} - \frac{D_j^0}{m^0_j}. \]  

Subsequently two simplifications are made. First, we assume that the difference between the terms related to the diffusive fluxes is negligible compared to the first term on the right-hand side. This means that Eq. (21) reduces to

\[ \Delta Z_j(s) \approx -\frac{1}{m} \int_{s_u}^{s} \rho K(Z_j - Z_{j,u}) \, ds. \]  

This assumption is correct if s is taken far away from the flame front where the diffusive fluxes D_j are zero. This was the case in the original work by de Goey and ten Thije Boonkkamp, but in this paper ΔZ_j will be evaluated at the inner layer where the diffusive fluxes are not small. The simplification also holds for weakly stretched flames for which the last two terms in Eq. (21) are the same. Second, we simplify Eq. (22) by assuming that K = constant and small and that all the values in Eq. (22) may be replaced by their unstretched counterparts. This results in

\[ \Delta Z_j(s) \approx -\frac{K}{m^0_j} \int_{s_u}^{s} \rho^0 (Z_j^0 - Z_{j,u}) \, ds. \]  

The same approach can be followed for the enthalpy changes, which yields

\[ \Delta h(s) \approx -\frac{K}{m^0} \int_{s_u}^{s} \rho^0 (h^0 - h_{u}) \, ds. \]
Both simplifications assume weak stretch and are valid when the stretch rates are sufficiently small. The effect of the different simplifications and the accuracy of expressions (23) and (24) to predict the values of the enthalpy change and the element mass fraction changes at the inner layer of stretched flames is studied here. In the theory of de Goey and ten Thije Boonkkamp, the equilibrium point at the burnt flame boundary ($s \to \infty$) was used in the expressions for $\Delta h$ and $\Delta Z_j$, where the fluxes $D_j$ are zero by definition. As noted, this is not the case at the inner layer where the most important chemical effects take place and where the mass burning velocity is evaluated, so that the last two terms in Eq. (21) do not cancel by definition.

The accuracy of the model can be seen in Fig. 1, in which profiles of $\Delta h$ and $\Delta Z_j$ are shown for a methane flame ($\phi = 0.8$). These profiles are taken from a numerical solution of the flamelet Eqs. (1)–(3) with a constant stretch rate $K = 100 \, \text{1/s}$. Similar plots were made for methane ($\phi = 0.9$), ethane, and propane. These figures are shown in Appendix A (Figs. 6–8). The local values of the enthalpy change and the element mass fractions changes are plotted versus the position $s$. The $s$ value of zero corresponds to the inner layer in the flame front, defined as the position where the heat release reaches a maximum. Negative positions are located on the unburnt side of the flame front, while positive positions refer to positions in the burnt gases of the flame. The position is scaled with the flame thickness $\delta_0$ of the unstretched flame. The differences between the numerical solution and the weak stretch theory are largest around the inner layer. At the burnt and unburnt side this difference reduces. The small remaining difference is caused by the finite stretch rate in the numerical solution which causes $Z_j \neq Z_j^0$ and vanishes if $K \to 0$. This means that neglecting the diffusive flux terms holds for the burnt and unburnt side, because the fluxes reduce to zero at the unburnt and burnt side of the flame. Around the inner layer of the flame the diffusive fluxes are quite large, which makes the inner layer a position, which is sensitive to the assumption of equal diffusive fluxes.
We also show the effect of the different simplifications made as a function of stretch in Fig. 2. In this figure $\Delta Z_C$ at the inner layer and at the burnt side is shown as a function of dimensionless stretch rate for methane ($\phi = 0.8$). It can be seen that for the burnt position, the enthalpy difference of the numerical solution and the solution with the assumption of equal diffusive fluxes are the same. This can be explained by the fact that diffusive fluxes are zero at the burnt side and thus the difference in diffusive fluxes is zero by definition. The weak stretch assumption introduces a deviation from the numerical solution, because the stretched flame structure and the unstretched flame structure are not exactly equal. This error grows with increasing $K$. At the inner layer position the assumption of equal diffusive flux terms causes an error additional to the error introduced by the weak stretch assumption. In fact, this is the main error at the inner layer, while at the burnt side it is the other way around. This is because the flux terms are zero at the burnt side, but the integral includes a much larger part of the difference between $Z_j$ and $Z_j^0$ than for the inner layer. In the next section the consequences of these deviations for the prediction of the mass burning rate will be shown. Furthermore, it must be noted that $\Delta Z_C$ is approximately 25% higher at the burnt side than at the inner layer. To show that this deviation results in a less accurate prediction of the mass burning rate, in Fig. 3 we have added predictions using values for $\Delta Z_j$ and $\Delta h$ at the burnt side of the flame. We can see that the predictions using numerical values of $\Delta Z_{j,b}$ and $\Delta h_b$ are less accurate for all flames in the weak stretch region. When the stretch rate increases the weak stretch assumption makes the model predictions less accurate.

5. Accuracy of the model for $m_{\text{fl}}$

Finally, the model for the mass burning rate is compared to the numerical solution in Fig. 3 for different fuels. The scaled mass burning rate is displayed as a function of the Karlovitz integral (scaled with the constant Lewis number), which is defined in Eq. (14). For all types of flames the flame stretch rates vary from $K = 0$ to $K = 300 \text{ s}^{-1}$. The Karlovitz integral is always smaller than 0.1. Three sets of theoretical results are displayed: one set for which the numerical values for $\Delta h$ and $\Delta Z_j$ at the inner layer are used, another set for which expressions (23) and (24) are used and one set with values for $\Delta h$ and $\Delta Z_j$ numerically evaluated at the equilibrium. This is done to evaluate the influence of using a weak stretch approach in the computation of $\Delta h$ and $\Delta Z_j$ on the final result and also to show that the inner layer is a better position to evaluate $\Delta h$ and $\Delta Z_j$ to make an accurate prediction of $m_{\text{fl}}$. It can be seen that the model is quite accurate, especially for weak stretch ($K_{\gamma,\text{il}} < 0.01$). In this range the predicted values are almost the same as the numerical values. For example, for a stretch rate of $K = 100 \text{ s}^{-1}$ the relative error is 4% for propane. The relative error in the other cases is smaller. For higher flame stretch rates the predictions become less accurate, due to deviations from the weak-stretch limit. The higher stretch rates cause larger differences between the stretched variables and the unstretched ones.

Markstein numbers are determined by taking the slope of the profiles displayed in Fig. 3 at $K_{\gamma,\text{il}} = 0$. This is done for the numerical and theoretical results for each type of flame. The results are presented in Table 2. The difference between the values displayed
Fig. 3. Mass burning rate $m_{il}$ scaled with $m_{0il}$ as function of Karlovitz number $Ka_{Yil}/Le_{Y}$. Full line, numerical solution; dashed line, $Le_{i} = 1$; symbols, weak stretch theory using numerical results for $\Delta h$ and $\Delta Z_{j}$ at inner layer ($\times$) and using Eqs. (23) and (24) at inner layer ($\circ$) and weak stretch theory using numerical results for $\Delta h$ and $\Delta Z_{j}$ at equilibrium ($\Delta$).

Table 2

<table>
<thead>
<tr>
<th></th>
<th>Methane $\phi = 0.8$</th>
<th>Ethane $\phi = 0.8$</th>
<th>Propane $\phi = 0.8$</th>
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</thead>
<tbody>
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<td>Numerical simulation</td>
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<td>3.03</td>
</tr>
<tr>
<td>Weak stretch theory</td>
<td>1.39</td>
<td>2.55</td>
<td>3.38</td>
</tr>
<tr>
<td>$\Delta h$ and $\Delta Z_{j}$ from numerical simulation</td>
<td>1.27</td>
<td>2.34</td>
<td>3.01</td>
</tr>
<tr>
<td>$\Delta h$ and $\Delta Z_{j}$ from Eqs. (23) and (24)</td>
<td>1.14</td>
<td>2.41</td>
<td>2.96</td>
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</tbody>
</table>

in Table 2 and the slopes in Fig. 3 is caused by the scaling factor $Le_{Y}$. The following definition is used to determine the Markstein number $M$:

$$\frac{m_{il}}{m_{0il}} = 1 - M \frac{Ka_{Yil}}{Le_{Y}}. \quad (25)$$

As in Fig. 3, Markstein numbers evaluated using numerical results for $\Delta h$ and $\Delta Z_{j}$ are also presented.

Table 2 confirms the results that were found in Fig. 3. The difference between the Markstein numbers found from the numerical simulations and the theoretical predictions are a good measure for the inaccuracy of the model. For ethane and propane the difference is smaller than 5%. However, the predictions for methane (difference of 20%) are less accurate than the predictions for ethane and propane. To understand the reason for this, let us investigate the magnitude of the different contributions $P_{h}$ and $P_{j}$ to the value of the mass burning rate. These terms are displayed in Fig. 4 for methane ($\phi = 0.8$) and propane to indicate the difference between these two fuels. The figures for methane ($\phi = 0.9$) and ethane are displayed in Appendix A (Fig. 9). Note that the different contributions of the enthalpy and elements have the same sign in lean ethane/air and propane/air flames, while they have opposite signs in both methane/air flames.
In the methane/air mixtures the different contributions partly cancel, which results in a relatively small overall preferential diffusion effect and therefore a larger relative error. This also explains the higher accuracy in ethane and propane flames investigated in this study. Figs. 4 and 9 show that the preferential diffusion effects for ethane as well as propane are larger than the preferential diffusion effects of the methane flames, as is commonly known.

Table 3 represents the slopes \( \partial \mathcal{P} / \partial (\text{Ka}_{Y;i,l} / \text{Le}_Y) \) of the different curves in Figs. 4 and 9 at \( \text{Ka}_{Y;i,l} = 0 \) for the different cases. Combining Eqs. (13) and (25) yields the relation between the sum of the slopes represented in Table 3 and the Markstein numbers in Table 2:

\[
\mathcal{M} = 1 - \text{Le}_Y \frac{\partial \mathcal{P}_h}{\partial \text{Ka}_{Y;i,l}} - \sum_{j=1}^{N_i-1} \text{Le}_Y \frac{\partial \mathcal{P}_j}{\partial \text{Ka}_{Y;i,l}}. \tag{26}
\]

The constant of 1 corresponds to the slope caused by the pure stretch term \( \text{Ka}_{Y;i,l} / \text{Le}_Y \), which is incorporated in Table 2 but not in Table 3. The same trend, i.e., cancelation of different terms, can also be seen in the individual Lewis numbers of the fuels; the Lewis number of methane is closer to one than the Lewis numbers of ethane and propane. In conclusion, a neutral preferential diffusive behavior is observed for lean methane/air flames, as in most other studies, and the influence of preferential diffusion increases with the fuel mass. A quantitative comparison of Markstein numbers with other values in the literature is not performed since different definitions are used by different authors. In addition it should be realized that in most other studies the value of the Markstein number depends on the position in the flame, which makes a quantitative comparison even more difficult.

Not only the Lewis number of the lean species, here the fuel, has a significant influence on the mass burning rate. The different preferential diffusion contributions can also be written in terms of species and temperature,

\[
\frac{m_{il}}{m_{il}^0} = 1 - \frac{\text{Ka}_{Y,i,l}}{\text{Le}_Y} \sum_{i=1}^{N_i} Q_i + Q_T, \tag{27}
\]

in which \( Q_i \) and \( Q_T \) are defined as

\[
Q_i = \left( c_h h_i^{\text{form}} + c_C W_{i,C} + c_O W_{i,O} + c_H W_{i,H} \right) \times (Y_{i,i,l} - Y_{i,l}^0), \tag{28}
\]

\[
Q_T = c_h (h_{il} - h_{il}^0) - \sum_{i=1}^{N_i} c_h h_i^{\text{form}} (Y_{i,i,l} - Y_{i,l,i}^0), \tag{29}
\]
with \( h_i^\text{form} \) the formation enthalpy of species \( i \). Contributions of the most important species (a total contribution of 90% or more) are shown in Figs. 5 and 10, where the contributions of the different species are shown separately as a function of the Karlovitz number. Also, Table 4 represents the values of the slopes \( \partial Q_i / \partial (K_{\text{Y,il}}/L_{\text{Y}}) \) at \( K_{\text{Y,il}} = 0 \) for the different cases. In this table the species and temperature are sorted in order of decreasing contribution. The contributions of the seven most important species are given, which corresponds to approximately 90% of the sum of the absolute values of all contributions. It can be seen that taking the preferential diffusion effect of the lean (fuel) species only is an incomplete approach. This confirms the results of the study by Mishra et al. [21], who showed that many intermediate species play an important role and that full chemistry is needed to predict stretch effects. Note that the order of the contributions is different for the different fuels and that the contribution of the fuel is not the maximum contribution. It becomes clear that several other species have an important influence on the mass burning rate. More specifically, the sum of the contributions of temperature, \( \text{CO}_2 \), \( \text{CO} \), \( \text{H}_2 \), and \( \text{H}_2\text{O} \) represents 72, 73, 78, and 70%, respectively, of the total contribution for the flames studied here. For ethane and propane the intermediates \( \text{H}, \text{C}_2\text{H}_4 \), and \( \text{C}_3\text{H}_6 \) also play an important role. The effect of these species can only be seen when detailed chemistry and multiple-species transport models are used. These results cannot be found by taking one-step chemistry and only one Lewis number.

### Table 4

<table>
<thead>
<tr>
<th>Contribution</th>
<th>( T )</th>
<th>( \text{CO} )</th>
<th>( \text{CO}_2 )</th>
<th>( \text{H}_2 )</th>
<th>( \text{O}_2 )</th>
<th>( \text{H}_2\text{O} )</th>
<th>( \text{CH}_4 )</th>
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</thead>
<tbody>
<tr>
<td>Slope</td>
<td>(-0.11)</td>
<td>(-0.10)</td>
<td>(-0.09)</td>
<td>(-0.06)</td>
<td>(0.04)</td>
<td>(-0.03)</td>
<td>(-0.02)</td>
</tr>
<tr>
<td>Cumulative</td>
<td>0.22</td>
<td>0.43</td>
<td>0.61</td>
<td>0.73</td>
<td>0.81</td>
<td>0.88</td>
<td>0.92</td>
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### \( \text{CH}_4, \phi = 0.9 \)

<table>
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<tr>
<th>Contribution</th>
<th>( T )</th>
<th>( \text{CO} )</th>
<th>( \text{H}_2 )</th>
<th>( \text{O}_2 )</th>
<th>( \text{H}_2\text{O} )</th>
<th>( \text{H} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Slope</td>
<td>(-0.11)</td>
<td>(-0.11)</td>
<td>(-0.10)</td>
<td>(-0.08)</td>
<td>(-0.07)</td>
<td>0.06</td>
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<tr>
<td>Cumulative</td>
<td>0.18</td>
<td>0.35</td>
<td>0.51</td>
<td>0.64</td>
<td>0.75</td>
<td>0.84</td>
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</table>

### \( \text{C}_2\text{H}_6, \phi = 0.8 \)

<table>
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<tr>
<th>Contribution</th>
<th>( T )</th>
<th>( \text{CO} )</th>
<th>( \text{H}_2 )</th>
<th>( \text{CO}_2 )</th>
<th>( \text{H}_2\text{O} )</th>
<th>( \text{C}_2\text{H}_4 )</th>
<th>( \text{H} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Slope</td>
<td>(-0.78)</td>
<td>(-0.51)</td>
<td>(-0.30)</td>
<td>(-0.25)</td>
<td>(0.18)</td>
<td>(-0.13)</td>
<td>(-0.09)</td>
</tr>
<tr>
<td>Cumulative</td>
<td>0.30</td>
<td>0.50</td>
<td>0.61</td>
<td>0.71</td>
<td>0.78</td>
<td>0.83</td>
<td>0.86</td>
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</table>

### \( \text{C}_3\text{H}_8, \phi = 0.8 \)

<table>
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<th>Contribution</th>
<th>( T )</th>
<th>( \text{CO} )</th>
<th>( \text{H}_2 )</th>
<th>( \text{C}_3\text{H}_6 )</th>
<th>( \text{CO}_2 )</th>
<th>( \text{C}_2\text{H}_4 )</th>
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<tbody>
<tr>
<td>Slope</td>
<td>(-1.51)</td>
<td>(-1.09)</td>
<td>(-0.48)</td>
<td>(-0.38)</td>
<td>(-0.22)</td>
<td>(-0.16)</td>
<td>(-0.15)</td>
</tr>
<tr>
<td>Cumulative</td>
<td>0.33</td>
<td>0.56</td>
<td>0.66</td>
<td>0.75</td>
<td>0.79</td>
<td>0.83</td>
<td>0.86</td>
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6. Conclusions

The model as introduced by de Goey and ten Thije Boonkkamp [9], modified in this work, has proven to provide an accurate basis to predict the mass burning rate of planar stretched flames with nonunit Lewis numbers in the weak stretch limit. Detailed chemistry and multispecies transport models were used with success to set up this quantitative flame stretch model. Several figures display the accuracy of this model. This new model includes more physics, like multispecies transport and the formation and destruction of radicals and intermediates. The assumption of equal...
Diffusive fluxes in unstretched and weakly stretched flames causes relatively small deviations in the predictive capacity of the theory. Furthermore, the model shows that the mass burning rate can be modeled accurately by considering the dependence of the mass burning rate on the element mass fraction and the enthalpy only. Accuracy might even be increased when more independent variables are included. It is interesting to see that the sensitivity coefficients for changes in mixture composition on the mass burning rate of the different fuels and flames investigated in this study have the same order of magnitude. It requires more research to determine the physical background of this phenomenon. Different contributions of the preferential diffusion terms to the mass burning rate are also analyzed, expressed in terms of enthalpy and element mass fractions as well as in terms of chemical species and temperature. From the last analysis it seems that only taking the Lewis number of the lean species is an incomplete approach since there is a number of other species (and temperature) that have a significant contribution. Mishra et al. [21] came to the same conclusion in their study. At the moment it is not clear which (combination of) Lewis number(s) has the most predictive value. Finally, it has been shown that the mass burning rate of methane/air flames is more difficult to predict than the mass burning rate of heavier fuels, because cancelation of contributions takes place in the case of methane. The approach presented in this paper can be used to identify stretch and preferential diffusion effects for an arbitrary fuel. The sensitivity for changes in the enthalpy and elemental composition must be evaluated numerically first. The magnitude of the enthalpy change and element mass fraction

Fig. 6. $\Delta h$ and $\Delta Z_j$ as a function of position ($K = 100 \text{ m/s}$) for methane ($\phi = 0.9$). Full line, numerical solution; dashed line, weak stretch theory. (a) $\Delta h$, (b) $\Delta Z_C$, (c) $\Delta Z_O$, (d) $\Delta Z_H$. 
changes can be computed from the models presented in the fourth section. Combining these two inputs and incorporating the Karlovitz integral gives good results in describing the mass burning rate. Future work will focus on the effects of strong stretch in combination with preferential diffusion on the mass burning rate. This could make it possible to describe the mass burning rate of strongly stretched flames without solving the complete flame structure.

Appendix A

Figs. 6, 7, and 8 show the change of enthalpy and element mass fractions as a function of position for methane ($\phi = 0.9$), ethane, and propane, respectively. Fig. 9 displays the contributions of preferential diffusion terms expressed in enthalpy and element mass fractions for methane ($\phi = 0.9$) and ethane. Fig. 10 shows the contributions of preferential diffusion terms expressed in temperature and species for methane ($\phi = 0.9$), ethane, and propane.

References

Fig. 8. $\Delta h$ and $\Delta Z_j$ as a function of position ($K = 100 \, 1/s$) for propane. Full line, numerical solution; dashed line, weak stretch theory. (a) $\Delta h$, (b) $\Delta Z_C$, (c) $\Delta Z_O$, (d) $\Delta Z_H$.

Fig. 9. Contributions of preferential diffusion terms expressed in enthalpy and element mass fractions for methane ($\phi = 0.9$) and ethane. Full line, numerical solution; symbols, weak stretch theory.
Fig. 10. Contributions of preferential diffusion terms expressed in temperature and species for (a) methane ($\phi = 0.9$), (b) ethane and (c) propane. With in figure (a), (×) CO$_2$, (○) CO, (⋆) temperature, (□) O$_2$, (△) H$_2$, (▽) H$_2$O, (★) H; in figure (b), (×) temperature, (○) CO, (⋆) H$_2$, (□) CO$_2$, (△) H$_2$O, (▽) C$_2$H$_4$, (★) H, (+=) CH$_3$; in figure (c), (×) temperature, (○) CO, (⋆) H$_2$, (□) C$_3$H$_6$, (△) CO$_2$, (▽) C$_2$H$_4$, (★) H, (+) O$_2$.