Behavior of Preheated Premixed Flames at Lean Conditions

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The ultra-lean combustion regime extends below the conventional lean flammability limit, where viable flames require substantial preheating of the reactant mixture. A large number of simulations of adiabatic laminar flames with detailed reaction kinetics and pseudo-randomly distributed inlet temperatures ($T_{in} = 300–1000$ K) and equivalence ratios ($\phi = 0.12–1.0$) is used to investigate fundamental aspects of lean and ultra-lean combustion of methane/air mixtures. Common features of flame characteristics are studied as a function of inlet temperature, equivalence ratio, mass flux and peak temperatures, which are used to parameterize the solution space. Holding individual parameters constant, a comparison of flame structures reveals that a characteristic temperature at the transition from preheating zone to the active reaction zone is reliably predicted by the local mass flux. Another characteristic temperature marking complete reaction of the fuel is predicted by the peak temperature. Overall, findings demonstrate that a parameterization using initial temperature and/or equivalence ratio fails to predict parameters of the inner reaction zone. Yet, results show that key features within the flame structure of premixed combustion are essentially unchanged throughout the lean and ultra-lean regime, where burning rates determine the temperature at which the preheating layer transitions into the active reaction zone. It is shown that peak temperatures determine characteristics of the active reaction zone irrespective of the mixture stoichiometry.

1 Introduction

Forthcoming combustion technologies employ various means for pollutant reduction, where lean premixed combustion is particularly promising [1]. Predictive models of reacting flows represent valuable tools for the development of modern combustion systems. In addition to the accuracy of the computational fluid dynamics (CFD) scheme, the quality of predictions is critically affected by the fidelity of the chemistry model. An accurate combustion model gives crucial information on the efficiency of a combustion process allowing the researcher to track the sources of deficiencies. Detailed mechanisms typically involve hundreds of species and thousands of reactions and contain comprehensive information regarding chemistry. As the computational cost scales with the number of conserved scalars, large-scale simulations using detailed mechanisms are generally not tractable. Due to the inherent computational expenses of detailed chemistry, considerable effort has been put into the development of methods to reduce the dimensionality of combustion processes. To perform dimensionality reduction, two main approaches have been pursued [2]: 1) mechanism reduction and 2) state-space parameterization.

In mechanism reduction, the chemical mechanism is modified to reduce the number of species and its stiffness. Conventional reduction techniques are based on the observation that due to very fast
chemical processes in combustion problems, many chemical species and reactions are in quasi-steady state or partial equilibrium, and algebraic relations are used to reduce the computational cost. In general, a vast knowledge of the chemical kinetics is required to decide which species can be assumed to be in steady state. While some automated methods for chemistry reduction have been proposed [3], resulting mechanisms are still rather large, and are typically not capable of modeling all regions within the flame structure.

In state-space parameterization, results of 1D simulations with detailed chemistry are used to span a manifold in which all properties are tabulated based on a small set of parameters that span a manifold. As the resulting chemistry tabulation is based on information obtained from detailed reaction mechanisms, full chemistry is implicit to simulation results. A main concern of this approach is a suitable parameterization of the flame manifold, where number and definition of parameters, as well as construction and implementation of manifolds in an actual combustion simulation have been addressed by a wide range of models in the literature. Steady Laminar Flamelet Method (SLFM) [4], Computational Singular Perturbation (CSP) [5], Intrinsic Low Dimensional Manifolds (ILDM) [6], flamelet based models [7, 8, 10, 11, 12], and more recently, the use of Principal Component Analysis (PCA) in combustion [2, 13, 14] are examples of models based on state-space parameterization.

Within state-space models, CSP and ILDM are based on the observation that a typical combustion system contains many chemical processes with a much smaller time scale than the flow time scales. In CSP, fast time scales are identified dynamically, i.e. the number and identity of the steady-state species may vary during the combustion process. Although this technique is very accurate, its applicability to complex simulations is questionable due to its high computational cost [5]. ILDM identifies the fast chemical processes by a local eigenvalue analysis of the chemical source term. ILDM has been successfully applied to many combustion simulations [15]. However, complex fuels and low-temperature kinetics require a large number of controlling variables for an accurate prediction of the burning velocity.

An alternative approach in state-space modeling uses detailed information of flame structures that are tabulated in terms of suitable parameters. The idea of considering a multidimensional manifold of one-dimensional flames/flamelets [9], has been applied to a wide range of predictive models such as Flamelet Progress Variable (FPV), Flamelet-Generated Manifolds (FGM), and Multidimensional Flamelet-Generated Manifold (MFM), among others. In FPV, chemistry is tabulated based on a progress variable, which is typically defined in terms of a linear combination of species mass fractions [12, 7]; additional parameters for chemistry tabulation capture diffusion and stretch effects to adjust to local conditions. FGM, introduced by van Oijen [10], expands the flamelet concept to multi-dimensional manifolds by including local elemental composition and enthalpy in addition to the progress variable in the tabulation. MFM is a closely related flamelet-based model that addresses processes observed in partially premixed combustion, where interactions between zones with different stoichiometry are captured by multi-dimensional manifolds [8].

Chemistry manifolds based on PCA seek to systematically identify a reduced set of principal components that describe the thermochemical state of a system [2, 13]. While PCA is most commonly used to extract common features from detailed combustion simulations, modeling approaches that use suitable transport equations for PC’s have been proposed [14].
In any of the state-space parameterization models, the main concern is whether the controlling variables are capable of capturing the flame characteristics. In the present work, a large number of simulations of lean premixed $\text{CH}_4$/air flames is used to investigate combustion characteristics over a large range of mixture stoichiometries and unburned temperatures. Detailed simulations involve detailed reaction kinetics and pseudo-randomly distributed inlet temperatures ($T_{in} = 300 - 1000 \text{ K}$) and equivalence ratios ($\phi = 0.12 - 1.0$). Within the flame structure, specific characteristics are investigated in terms of species concentrations, their respective net production rates, heat release and net reaction rates. A suitable parameterization for a flame manifold is sought, based on an analysis of cases sharing common normalized equivalence ratio $\Phi = \frac{\phi}{1+\phi}$, unburnt temperature $T_{in}$, laminar burning flux $\dot{m}_L$ and adiabatic flame temperature $T_{ad}$. Based on a suitable parameterization, the generalization from one-dimensional results to arbitrary flow fields is discussed.

2 Methodology

The overarching goal of the study is to identify flame characteristics that are shared by a large number of premixed flames with varying mixture stoichiometry and unburned temperature. Results from a precursor study [16] indicate that characteristic temperatures within the inner layer of the flame sheet remain essentially unchanged for a wide range of operating conditions. For a detailed analysis of the characteristics of the flame structure, 697 one-dimensional test cases are created, where unburned temperature $T_{in}$ and normalized equivalence ratio $\Phi$ follow a pseudo-random Sobol sequence. Simulations are obtained and using GRI-Mech 3.0 [17] and CANTERA [18]. An overview of the test cases is presented in Figure 1, which illustrates contours of laminar burning flux $\dot{m}_L$ and adiabatic flame temperature $T_{ad}$ as a function of $T_{in}$ and $\Phi$.

In order to assess the flame characteristics based on physical locations for the entire data set, the following functions characterizing the flame structure are defined: (a) species concentrations, (b) their respective net production rates $\dot{\omega}$, (c) heat release $\dot{H}$, and (d) net reaction rates, defined by the law of mass action as the product of reactant concentrations and specific reaction rate constants at a given temperature [19]. In each case, characteristic temperatures $T_{i,j}^*$ are cataloged for the
Figure 2: Definition of characteristic temperatures $T_{i,j}^*$ based on physical locations of extrema of functions values and derivatives. Different symbols are used to mark minima/maxima of function values ($T_{0,j}^*/\circ$), first derivatives ($T_{0,j}^*/\times$) and second derivatives ($T_{0,j}^*/+$), where derivatives are taken with respect to position.

$j^{th}$ minimum /maximum of the function itself ($i = 0$), as well as first ($i = 1$) and second ($i = 2$) derivatives of the function with respect to the position within the flame. Figure 2 illustrates the definition for characteristic temperature of selected major species within the flame structure. Due to the rapid temperature increase, a presentation of the flame structure as a function of local temperature yields improved insights into the magnitude of characteristic temperatures within a reaction zone. It is noted that characteristic temperatures are defined for physical locations within the flame structure (Figure 2 - left), and derivatives based on temperature (Figure 2 - right) are not considered. Also, the strong curvature of $df/dT$ at low temperatures is an effect of the Lewis number.

3 Results and Discussion

3.1 Parameterization of Flame Manifolds

A first comparison of flame structures is obtained for sets of cases that share common parameters that either define inlet conditions or constitute major results of flame simulations: normalized equivalence ratio $\Phi$, unburned temperature $T_{in}$, laminar burning flux $\dot{m}_L$ and adiabatic flame tem-
Figure 3: Flame structures for simulations with comparable stoichiometry $\Phi$, inlet temperature $T_{in}$, laminar burning flux $\dot{m}_L$, and adiabatic flame temperature $T_{ad}$ (from left to right). Flame structures are comparable for cases where characteristic temperatures are shared, i.e. markers (◦/×/+ ) align vertically.

The temperature $T_{ad}$. Figure 3 illustrates results, where each inset shows a comparison of specific species mass fraction profiles for comparable parameters. Results illustrate that cases with shared mixture stoichiometry ($\Phi$) display dissimilar combustion characteristics. Likewise, the inlet temperature contains little information, despite a seeming collapse of $O_2$ and $H_2O$ concentrations that is due to a shared starting point in the graph. However, results for cases with shared laminar burning flux $\dot{m}_L$ reveal that the steepest gradient of major species occurs at the same temperature, which indi-
Figure 4: Comparison of different parameterizations for characteristic temperatures for CO concentrations, calculated for 697 test cases as shown in Figure 1. \( T^*_{0,0} \) (CO) mark temperatures where CO concentrations peak, whereas \( T^*_{1,j} \) (CO) and \( T^*_{2,j} \) (CO) mark temperatures with largest gradient and curvature, respectively.

Indicates a significant transition within the flame structure; specifically, this behavior is attributed to a transition from the preheat zone to the active reaction zone. Cases with comparable adiabatic flame temperatures reveal common traits for the high-temperature portion of the species profiles, which is attributed to fast chemistry within the active reaction zone governed by a reaction-diffusion process.

In order to reveal shared flame characteristics, characteristic temperature are calculated for the entire data set of 697 cases shown in Figure 1. The four insets of Figure 4 show results for characteristic temperatures of CO concentrations of all simulations parameterized by \( \Phi \), \( T_{in} \), \( \dot{m}_L \) and \( T_{ad} \), respectively. Results further illustrate that information on either of the two inlet conditions (i.e. \( \Phi \), \( T_{in} \)) alone is meaningless for a universal parameterization of the flame structure, despite the fact that they constitute the most convenient parameterization. In comparison, a parameterization based on \( \dot{m}_L \) shows a good collapse of the steepest gradient in CO concentrations, \( T^*_{1,0} \) (CO), which is attributed to a transition from the convection-diffusion dominated preheating layer to the reaction-diffusivon dominated active reaction zone. A parameterization based on \( T_{ad} \) results in an almost perfect collapse of high-temperature characteristics of CO concentrations, which implies that the peak temperature is the singlemost important parameter characterizing processes within the active reaction zone. While this result is obtained by a purely empirical approach based on flame simulations with widely ranging stoichiometry and inlet conditions, it is consistent with conventional reaction kinetics, where reaction rates defined by Arrhenius kinetics are majorly impacted by local temperatures. In addition, the result illustrates that simplified qualitative modeling approaches (e.g. large-activation energy asymptotics [20]) capture essential physics.

One shortcoming of using profiles of species concentrations as a basis for the definition of characteristic temperatures is that they are subject to both transport processes and chemical reactions. Species production rates \( \dot{\omega} \) as well as heat release \( \dot{H} \), however, are due to chemical reactions only, which makes them well suited for a characterization of the active reaction zone. Figure 5 illustrates characteristic temperatures \( T^*_{i,j} (\dot{\omega}_s) \) for several significant species, where results are parameterized by \( T_{ad} \). The collapse of characteristic temperatures for comparable \( T_{ad} \) indicates that, except for a scaling factor, species production rates and heat release follow almost identical progressions.
This result implies that a reduced manifold for a description of chemical kinetics can be obtained, where source terms are tabulated based on two parameters: local temperature and the adiabatic temperature based on local conditions.

3.2 Generalization to Arbitrary Flow Fields

Results discussed so far are a strong indication that a generic flame structure does indeed exist, although preliminary findings are restricted to CH₄/air flames that are adiabatic and one-dimensional. It has been shown that characteristic temperature for net species production rates and heat release can be collapsed based on the adiabatic flame temperature ($T_{ad}$), which can be used to tabulate source terms as a function of local temperature and adiabatic temperature. Since this tabulation is based on the adiabatic flame temperature, a generalization of this procedure to arbitrary non-adiabatic and/or curved flames requires estimates based on local mass fractions, temperature and associated transport processes.

According to the first law of thermodynamics, the enthalpy of the burned products of a freely
Once the final enthalpy is known, the initial temperature of the unburned mixture can be obtained by balancing elemental species to form a suitable unburned mixture and equating enthalpies. For adiabatic temperatures can be easily obtained by thermal equilibrium, Figure 6 illustrates that estimates of the final (adiabatic) temperature based on local temperature and mole fraction alone, i.e. \( \hat{T}_{fl}(T, X_k) \), are inaccurate due to non-constant specific enthalpies.

Once the final enthalpy is known, the initial temperature of the unburned mixture can be obtained by balancing elemental species to form a suitable unburned mixture and equating enthalpies. For \( \text{CH}_4/\text{air} \) mixtures, reactants involve \( \text{CH}_4, \text{H}_2, \text{O}_2, \text{N}_2 \) and \( \text{Ar} \), where \( \text{H}_2 \) was added to accommodate deviations due to differential diffusion. The elemental species mass fractions \( Y_\alpha \) follow from tallying up elemental species within the mixture, and the original composition is estimated from a balance of elemental species. Estimates for the initial temperature based on local temperature and mole fractions alone, i.e. \( \hat{T}_{in}(T, X_k) \), are shown in Figure 6. Again, estimates are inaccurate due to non-constant specific enthalpies.

Accurate estimates of final and initial temperatures involve an improved estimate of the final enthalpy, which is obtained by integrating the energy equation from the local conditions to final conditions far downstream of the flame sheet of an adiabatic flame. In terms of enthalpy, the energy equation can be written as

\[
\rho \frac{Dh}{Dt} = \frac{Dp}{Dt} - \nabla \cdot \mathbf{q} - \sum_k \nabla \cdot (h_k \mathbf{j}_k) + \Phi
\]  

(1)

where \( h \) and \( \rho \) are specific enthalpy and density of the mixture, \( p \) is the pressure, \( \mathbf{q} \) is the heat flux, \( h_k \) and \( \mathbf{j}_k \) are specific enthalpy and mass flux of species \( k \), and \( \Phi \) is the viscous dissipation. Assuming constant pressure, neglecting viscous dissipation, and expanding the substantial derivatives for steady-state conditions, i.e. zero derivatives with respect to time or \( \partial_t(\cdot) = 0 \), the

\[ \hat{T}_{in}(T, X_k) \]
equation becomes
\[ \rho \mathbf{v} \cdot \nabla h = \nabla \cdot (\rho \mathbf{v} h) = -\nabla \cdot \mathbf{q} - \sum_k \nabla \cdot (h_k \mathbf{j}_k) \] (2)

where the first equality uses the chain rule of differentiation and the continuity equation at steady state. In the following, Eq. 2 is integrated over the volume of an adiabatic stream tube extending from local conditions to final conditions far downstream of the flame sheet, where all gradients vanish. Applying the divergence theorem
\[ \int_V \nabla \cdot (\rho \mathbf{v} h) \, dV = \oint_S \mathbf{n} \cdot (\mathbf{q} - \sum_k h_k \mathbf{j}_k) \, dS \]
and rearranging follows
\[ \dot{m} h_{fl} = \dot{m} h_{loc} - \mathbf{n} \cdot \mathbf{q}_{|loc} - \sum_k \mathbf{n} \cdot (h_k \mathbf{j}_k)_{|loc} \] (3)

where \( \dot{m} = |\mathbf{n} \cdot (\rho \mathbf{v})| \) = const along the stream tube, and subscripts loc and fl indicate local and final states, respectively. Assuming one-dimensional flame propagation and expanding \( \mathbf{q} \) and \( \mathbf{j}_k \) using expressions for thermal and mixture-averaged species diffusion, respectively [21], the specific enthalpy at the final state is estimated as
\[ h_{fl} = h_{loc} - \frac{1}{\dot{m}} \left[ \lambda \frac{dT}{dx} \bigg|_{loc} + \sum_k \rho h_k w_k D'_{km} \frac{dX_k}{dx} \bigg|_{loc} \right] \] (4)

where \( \lambda \) is the thermal conductivity, \( T \) is the temperature, \( W_k \) and \( \bar{W} \) are species and average molecular weight, \( D'_{km} \) is the mixture-averaged species diffusion coefficient, and \( X_k \) is the molar fraction of species \( k \). As shown in Figure 6, estimates for final and initial temperatures based on local temperature and mole fraction as well as heat and species fluxes, i.e. \( \hat{T}_{fl} (T, X_k, q, j_k) \) and \( \hat{T}_{in} (T, X_k, q, j_k) \), are somewhat improved, but still deviate from the actual simulated results.

The deviations are explained by non-constant elemental mass fractions throughout the flame structure, which, similar to non-constant enthalpies, are due to differential diffusion. Again, an improved approximation for the species mole/mass fractions at the final state can be obtained by integrating from local to final conditions. The conservation of elemental species is given by
\[ \rho \frac{DY_{\alpha}}{Dt} = \rho \partial_t Y_{\alpha} + \rho \mathbf{v} \cdot \nabla Y_{\alpha} = -\nabla \cdot \mathbf{j}_{\alpha} \] (5)

Again, assuming steady-state conditions and integrating over a stream tube from local conditions to the final conditions, the final elemental composition is estimated as
\[ Y_{\alpha,fl} = Y_{\alpha,loc} - j_{\alpha} / \dot{m} = Y_{\alpha,fl} - \frac{1}{\dot{m}} \sum_k n_{k,\alpha} W_{\alpha} \bar{W}_k j_k \] (6)

where \( n_{k,\alpha} \) is the number of atoms of elemental species \( \alpha \) in molecular species \( k \). The corrected elemental composition represents conditions both far downstream and far upstream of the flame sheet, which allows for estimates of both initial and final temperatures based on a representative unburned mixture composition. Figure 6 illustrates that estimates for final and initial temperatures based on local temperatures, mole fractions and fluxes of heat, molecular species, and elemental species, i.e. \( \hat{T}_{fl} (T, X_k, q, j_k, j_{\alpha}) \) and \( \hat{T}_{in} (T, X_k, q, j_k, j_{\alpha}) \) are accurate.

Using conservation of energy and species as well as thermodynamic equilibrium, it is possible to extrapolate from state and fluxes at a single point within a flame to a corresponding adiabatic
flame. Since $T_{\text{loc}}$ is known and both $T_{\text{in}}$ and $T_{\text{fl}}$ can be estimated, it is possible to extract the relative position within the flame based on data from a single point. Likewise, information of local species concentrations $X_{k,\text{loc}}$ and fluxes of elemental species can be used to infer both $X_{k,\text{in}}$ and $X_{k,\text{fl}}$. Since various formulations of progress variables for premixed flamelets are based on this information [22, 7], it is noted that progress variables corresponding to detailed numerical simulations with full chemistry can be calculated from a single point.

4 Conclusions

A comprehensive analysis of flame structures for a large number of one-dimensional simulations of premixed adiabatic CH$_4$/air flames demonstrates that essential flame characteristics are captured by two parameters: laminar burning mass flux $\dot{m}_L$ and adiabatic temperature $T_{\text{ad}}$. It was shown that a parameterization based on laminar burning mass flux $\dot{m}_L$ captures a significant transition within the flame structure, which is attributed to a transition from the convective-diffusive preheating layer to the reaction-diffusive dominated active reaction zone. A parameterization based on $T_{\text{ad}}$ reveals an almost perfect collapse of high temperature characteristics implying that the peak temperature is the singlemost important parameter characterizing processes within the active reaction zone. It was also shown that results based on production rates as a function of $T_{\text{ad}}$ capture the flame characteristics more promisingly than a comparable from concentrations. The collapse of features within the active reaction zone confirms the strength of the classical methods, in which solutions are parameterized based on the peak temperature. However, this method cannot resolve the flame structure using a global reaction for the chemistry. It is noted that a parameterization based on inlet conditions ($\Phi$ and $T_{\text{in}}$) does not lead to a collapse, indicating that a parameterization based on inlet conditions does not lead to manifolds with a minimized number of dimensions.

Results demonstrate that a manifold with minimized dimensions requires the adiabatic temperature $T_{\text{ad}}$. Thus, a generalization of one-dimensional results to arbitrary flow fields requires accurate estimates based on locally defined quantities, i.e. temperature, species, and associated fluxes. Since the thermochemical state at an individual point fully defines reaction processes, it is possible to relate an individual point within an arbitrary reacting flow to a corresponding premixed flame that shares the same local thermochemical state. Based on local information, it was shown that accurate estimates for adiabatic temperature and unburned temperature can be calculated based on path-integrated governing equations and thermodynamic equilibrium, which makes it possible to extract flamelet information a posteriori from detailed combustion simulations.

Findings from one-dimensional simulations suggest that chemistry tabulation based on manifolds spanned by $T_{\text{ad}}$ and $\dot{m}_L$ will result in a the dimension reduction for 2-d and 3-d combustion simulations as local thermochemical states within arbitrary flow fields can be related to corresponding one-dimensional premixed flames. In future work, it will be of interest to implement PCA models on production rates and heat release to determine a minimum set of principal components that allows for accurate estimates of adiabatic temperatures.
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References


