Analysis of Methane-Air Edge Flame Sturcture using CSP

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Abstract—We study the structure of a methane-air edge flame stabilized against an incoming mixing layer. The flame is computed using detailed chemical kinetics, and the analysis is based on computational singular perturbation theory. We focus on examination of the low-dimensional structure of the flame, analyzing the number of exhausted modes, along with the distribution of fast and active timescales. Results are used to enhance understanding of the flame, and the role of different chemical and transport processes in its observed structure.

I. INTRODUCTION

Edge flames can be encountered in many reacting flow configurations in which partial premixing of fuel and oxidizer occurs [1]–[7]. The idealized edge flame is composed of three distinct branches: a lean premixed branch, a rich premixed branch and a diffusion flame branch; hence it is also known as a triple or tribrachial flame. The two premixed branches form curved fronts behind which a diffusion flame develops and stabilizes. Symmetric, tribrachial, edge flame structure, as well as bibrachial and monobrachial edge flames have been observed, depending on the incoming mixture composition as well as the flow/mixing layer structure.

There is a large number of experimental, analytical and numerical studies of triple and edge flames [2], [8]–[13]. The reviews by Buckmaster [6] and Chung [7] provide a broad overview of the history and recent developments in the study of edge flames. Methane-air triple flames in particular have been the focus of numerous studies [4], [5], [14], [15].

Based on the literature, it is evident that detailed C_2 kinetics are necessary in modeling these flames. Takahashi *et al.* [16] studied methane-air edge flame structure using skeletal 17-species kinetics. The detailed chemical structure of a methane-air edge flame was also studied by Takahashi and Katta [17] using C_2 and C_1 chemistry mechanisms. Inclusion of C_2 kinetics was found to lead to an increase in the flame lift-off height. Use of a global single-step reaction mechanism was found to lead to significant differences in edge flame stability and structure, including a dominant rich premixed branch, in disagreement with the detailed chemistry predictions [18].

Walsh *et al.* [19] used a 26-species C_2 mechanism for computations of lifted methane-air diffusion flames. They generally found good agreement with experimental measurements, except at diluted fuel conditions. Ali and Daou [20] presented an analytical study of the effect of reversibility of chemical reactions on triple flames. Cho and Takita [21] presented computational studies of edge flames using detailed kinetics for both methaneair and propane-air mixtures. Briones *et al.* [22] conducted a numerical investigation of methane-air edge flames using detailed kinetics, with a focus on liftoff, stabilization, and blowout. Guo *et al.* [23] presented a numerical study of methane-air edge flames using a subset of GRImech3.0 [24] excluding Nitrogen chemistry. Najm *et al.* [25] studied the NO structure in a methane-air edge flame using GRImech3.0 [24], analyzing the four significant NO_x pathways.

In the present work we consider the methane-air edge flame of [25], and focus on the analysis of the flame structure using Computational Singular Perturbation (CSP) theory [26], [27]. CSP analysis enables identification of fast and slow reaction processes in chemical models, and the decoupling of fast-exhausted/dormant modes from the slow modes that drive the time evolution of the chemical system. The method relies on the identification of a suitable set of basis vectors that enable the decoupling of fast and slow processes. A leading-order approximation of these vectors is provided by the eigenvectors of the Jacobian of the chemical source term. There is an extensive literature on CSP and its utilization for analysis and reduction of chemically reacting flows [26]–[41].

We set up the methane-air edge flame stabilized against a prescribed uniform-velocity mixing layer using detailed chemistry (GRImech3.0 [24]) and mixture-averaged transport [42]. CSP analysis of the computed flame structure highlights the spatial variation of the fast and driving time scales and the number of exhausted modes over the edge flame, revealing significant internal structure, and identifying important associated chemical/transport processes. In the following, we outline the setup of the problem, then present the analysis of the edge flame using CSP. We finish with conclusions summarizing the main findings of the work.

II. PROBLEM SETUP

We consider a methane-air edge flame in twodimensions (2D) stabilized against an incoming mixing layer flowfield with a uniform inflow velocity profile. The computational model uses the low Mach number approximation, and employs GRImech3.0 [24] kinetics



Fig. 1. The two-dimensional spatial distribution of temperature, shown with superposed heat release rate and ξ -contours (a) and the dimension of the fast subspace M (b), shown with superposed w_{CH_4} and ξ -contours. The ξ -contours are drawn at $(0.9, 1.0, 1.1)\xi_{\mathrm{st}}$. This is a 512×1024 subset of the domain, with $x \in [0.4, 1.2]$ cm, $y \in [0, 1.6]$ cm.

and mixture-averaged transport [42]. The rectangular domain size is $1.6 \times 3.2 \text{ cm}^2$. The inflow is specified with a uniform velocity of 60 cm/sec and an analytically prescribed mixture-fraction profile, going from pure air on one side of the domain to pure CH₄ on the other [25]. The mixture fraction ξ is defined as in Bilger [43]. Note that ξ is zero in the air stream, and unity in the fuel stream. The stoichiometric value of the mixture fraction is $\xi = \xi_{st} = 0.055$ for the given fuel/air streams.

The global structure of the edge flame is illustrated in Figure 1(a). The inflow at the bottom edge of the domain is at 300 K, with the fuel stream on the right and the air stream on the left. The mixture fraction contours are drawn at $(0.9, 1.0, 1.1)\xi_{st}$. The flame temperature field is shown, exhibiting a fast rise from the cold reactants temperature to that of the hot combustion products across a narrow region where the primary edge flame reaction zone exists. The contours of the consumption rate of the fuel, w_{CH_4} , are superposed to highlight the curved premixed front structure, with clear rich and lean premixed branches. The region behind the premixed flame edge, extending along the central $\xi_{\rm st}$ line upwards through the domain, has a diffusion-flame structure, approximating the ideal one-dimensional non-premixed flame structure with downstream distance.

III. CSP ANALYSIS OF THE EDGE FLAME STRUCTURE

We recall that, with an N-dimensional ODE system, dy/dt = g, CSP employs a set of basis vectors/covectors, a_i/b_i , i = 1, ..., N, to decompose the source term into a sum of (ideally) decoupled modes $\{\mathfrak{M}_k\}_{k=1}^N$, given by $g = \sum_{r=1}^M a_r f^r + \sum_{s=M+1}^N a_s f^s$, where f^k is the amplitude of \mathfrak{M}_k . The modes are ordered from fastest to slowest, with associated time scales $\tau_1 < \cdots < \tau_N$. The first M (fast) modes, having sufficiently small amplitude according to chosen thresholds, are either "frozen", having no chemical activity, or "exhausted", involving



Fig. 2. M cut along different y levels



Fig. 3. WCH4 cut along different y levels

cancellations of opposed processes. The remaining N-M modes are slow, constrained to evolve along a slow manifold defined by the fast processes. In a PDE context, we identify the CSP basis vectors based on the chemical source term alone, however, the determination of M is based on the projection of the full right-hand-side on this basis. Finally, for any given mode k, the CSP Participation Index (PI) of any process i, \mathcal{P}_k^i , is a normalized measure of the importance of that process in this mode, where a process is defined as any of the forward/backward reactions in the system, or a convection/diffusion transport term for a given species.

With this cursory definition of terminology, we now examine the structure of the edge flame using CSP analysis. Figure 1(b) shows the spatial distribution of M, the dimension of the fast subspace, with superposed w_{CH_4} and ξ -contours. The detailed structure of the M field is complex, but some key features may be readily observed in this 2D plot. The primary reaction zone region, where w_{CH_4} is largest, exhibits generally the lowest M values, where only 3-8 modes are exhausted. Note however, the misalignment between the w_{CH_4} contours and the region with lowest M values in the downstream end of the rich edge flame branch. The central, diffusion flame region has the highest number of exhausted modes, with M > 30. At



Fig. 4. The spatial distribution of the fastest chemical timescale τ_1 (a), and the driving chemical time scale τ_{M+1} (b), in the edge flame, shown with superposed w_{CH_4} and mixture fraction contours. The ξ -contours and domain size are as those in Fig. 1. Note that the color map for the τ_{M+1} plot has been truncated at 1 μ sec for clarity.

the base of the edge flame, this high-M region coincides with the $\xi = \xi_{st} = 0.055$ line, shifting towards leaner mixtures mid-way up the domain. This is the region which is closest to equilibrium as the reaction products from the lean/rich sides of the edge flame come together.

Further, horizontal cuts of the domain at three ylocations of (0.3, 0.5, 0.8) cm, showing the spatial profiles of M and the chemical production rate of the fuel, $w_{\rm CH_4}$, at different elevations in the edge flame, are shown in Figures 2, and 3 respectively. The high M-plateau in the cold gas is evident in the 0.3 cm profile, with $M \sim 22$ for $x \sim 0.4, 1.2$ cm, and $w_{\rm CH_4} \sim 0$. As we approach regions of significant CH₄ consumption, with $x \sim 0.7, 0.93$ cm, we find largely monotonic decrease in M towards a minimum that roughly coincides with the peak consumption rate of the fuel. Going further, towards the domain centerline, M is found to increase again, with an eventual peak value at $x \sim 0.8$ cm. This picture is roughly similar at the other two y-elevations. The central peak is observed to shift towards the left, reflecting the shift observed in the 2D image in Fig. 1(b). Similarly, the locations of the minima in M are observed to shift away from the centerline, reflecting the increased width of the edge flame in Fig. 1(b). Further, these minima are broader reflecting the increased thickness of the reaction zone at these rich/lean locations. The minimum value of M also increases, indicating that less/more modes are active/exhausted in the premixed front nearer stoichiometric conditions. Finally, note that the deviation between the location of the M-minimum and peak fuel consumption in the rich region at higher y-elevations, as observed in Fig. 1(b).

Figure 4(a) shows the spatial distribution of the fastest chemical timescale τ_1 in the edge flame. The cold fuelrich region exhibits the fastest τ_1 , ~ 0.7 ns, while the curved preheat zone ahead of the primary edge flame reaction zone exhibits the slowest τ_1 , ~ 2.7 ns. In this zone, important low temperature flame processes are active. Specifically, this zone coincides with the regions of low temperature peak production rates of HO₂ and CH₃O, and associated processes. Thus, for example, the conversion of NO to NO₂ is peaked in this region, dominated by the forward rate of the reaction R₁₈₆: HO₂+NO = NO₂+OH. Here and in the following, we use the reaction numbers to identify the specific reaction in GRImech3.0 [24]. Moreover, we adopt the following convention, denoting the net, forward, and backward reaction *i* by R_i, F_i, and B_i respectively.

Along the domain inlet, and spanning the x-length of the domain, we find that the processes with the largest PI in mode 1, \mathcal{P}_1^i , are largely the convection and diffusion of O₂. Occasionally, *i.e.* at some x-locations, the diffusive fluxes of CH₄ dominate, but the far majority of points exhibit the stated dominance of O₂ transport fluxes in mode 1. Note that, in this region there is barely any chemical activity, in fact many modes are frozen, such that the only significant activity is the transport of reactants. Thus, the observed dominance of these transport processes in the PI of mode 1 (PI₁), indicating that only transport is significant along a_1 , is not surprising.

Looking at cuts at three elevations through the edge flame (0.3, 0.5, 0.8 cm), the PI results for mode 1 at all ylocations are as follows. The results are essentially similar on either side of the flame, in the fuel or air streams, as at the lower location near the domain inlet, with dominance of O_2 transport processes. As we enter the preheat zone on either side of the thin premixed flame zones, various transport processes gain dominance in narrow spatial regions, namely diffusion and convection of Temperature, H_2 , and H_2O , along with the diffusion and convection of CO on the air-side only. Going further into the preheat zone, a range of reactive processes exhibit narrow regions of dominance. These are: F_{97} : OH+CH₃ \Rightarrow CH₂(S)+H₂O, F_{125} : CH+O₂ \Rightarrow O+HCO, F_{130} : CH+CH₄ \Rightarrow H+C₂H₄, F_{142} : $CH_2(S)+N_2 \Rightarrow CH_2+N_2$, B_{126} : $CH+H_2 \Leftarrow H+CH_2$, and, in the air-side preheat zone only, F_{144} : CH₂(S)+O₂ \Rightarrow H+OH+CO. These reactions are part of the cascade of fast fuel-breakdown reactions. As for the region inside the edge flame, including on either side the two primary flames, the reaction R_{204} : NNH = N_2 + H, has the dominant participation index in mode 1 at all y-locations.

Moreover, recall that examining the CSP pointer for exhausted modes, allows the identification of the associated CSP radical species for that mode. In other words, this is the species whose fast consumption processes have high PIs, and therefore are key participants in the associated equilibrium and corresponding time scale of this mode. In the present case, and considering all *y*-locations, we find the following regarding mode 1. In the cold flow region outside the edge flame, the species pointed to by mode 1 is CH. Further, this mode is found to be either frozen or exhausted in different parts of this region. While the transport of O_2 or CH₄ dominates the PI, this is, as indicated above, because there is hardly any chemical activity. However, recall that the time scales τ_k result from the analysis of the *chemical* source term. Further,

the fact that this mode points to CH suggests that CH consumption reactions are relevant here in establishing τ_1 . In fact, looking at the most dominant PIs for *chemical* processes we find that reactions F_{125} : CH+O₂ \Rightarrow O+HCO (on the air side) and F_{130} : CH+CH₄ \Rightarrow H+C₂H₄ (on the fuel side), both CH consumption reactions, are generally among the highest amplitude PIs among the reactive processes in this region. Further, it is likely that the difference in τ_1 between the cold fuel and air streams, evident in Fig. 4(a), is a direct result of the dominance of one or the other of these reactions in each zone. In the preheat zones, where low-temperature chemistry is active, and mode 1 is exhausted, the associated CSP pointer points to $CH_2(S)$. This is consistent with the observed dominance of several reactions involving $CH_2(S)$ in this region in PI_1 , as observed above. On the other hand, inside the edge flame, NNH is the corresponding species pointed to by mode 1, again exhausted, which is again consistent with the above dominance of R_{204} in PI_1 in this region.

Consider next the spatial structure of the driving chemical time scale τ_{M+1} , shown in Figure 4(b). Recall that \mathfrak{M}_{M+1} is the fastest of the slow modes, such that the system evolves locally with time scale τ_{M+1} . We note first that the cold fuel/air regions, where largely only slow transport processes are active, exhibit a large $\tau_{M+1} \ge 1 \mu$ sec. Further, as we approach the primary flame, in the preheat region, a narrow region of alternating fast/slow activity is evident, before the broad region of fast chemical activity in the edge flame is reached. In this internal region, fastest chemical activity is evident in the primary premixed flame zone, with slower τ_{M+1} evident in the central region around ξ_{st} where chemical equilibrium is approached.

Let us examine the PIs for \mathfrak{M}_{M+1} along horizontal cuts. As for \mathfrak{M}_1 , the lack of chemical activity in the cold flow regions outside the edge flame highlights the action of transport fluxes. Here again, we find the transport of O_2 to dominate the PI. Among reactive processes, the reactions with the largest PIs (albeit small relative to transport) in this region are found to be F_{11} : O+CH₄ \Rightarrow OH+CH₃, F_{179} : N+O₂ \Rightarrow NO+O, F_{216} : HNO+O₂ \Rightarrow HO₂+NO, F_{291} : CH₂+O₂ \Rightarrow O+CH₂O, and B₂₀₇: NNH+O \Rightarrow OH+N₂. As we approach the flame edge, reaction fluxes become significant, dominating the role of transport in PI_{M+1} . In the preheat zone, reactions such as F_{119} : HO₂+CH₃ \Rightarrow OH+CH₃O, and F_{98} : OH+CH₄ \Rightarrow CH₃+H₂O, as well as F_{170} : $CH_3O+O_2 \Rightarrow HO_2+CH_2O$ are found to be dominant. This is a reflection of the dominant role of these reactions in the fuel breakdown chemistry on the reactants side of the premixed flame.

Finally, we note that the detailed structure inside the flame is quite complex. Consider in particular the region with minimum M, in the vicinity of the primary premixed flame reaction zone, shown in Figs. 1(b) and 2. In this region, and focusing on a cut at y = 0.3 cm for illustration, we find that reactions with dominant PI_{M+1} in the lean branch include F_{97} : OH+CH₃ \Rightarrow CH₂(S)+H₂O,

F₆₈: H+CH₃OH ⇒ CH₂OH+H₂, F₁₆₉: CH₂OH+O₂ ⇒ HO₂+CH₂O, and F₁₂₆: CH+H₂ ⇒ H+CH₂. On the other hand, in the rich branch, the following reactions dominate this mode, F₁₂₂: C+O₂ ⇒ O+CO, F₅₈: H+CH₂O ⇒ HCO+H₂, F₁₆₈: HCO+O₂ ⇒ HO₂+CO, along with B₁₂₆. A detailed study of the internal structure of the premixed flame primary reaction zone is greatly informed by these specific findings, identifying the reactive processes controlling the chemical evolution of the system in each internal flame layer. Similar information is equally useful in other internal regions of the edge flame.

IV. CONCLUSIONS

We have studied the structure of a methane-air edge flame using CSP analysis. Results identified the structure of the flame at different spatial locations, illustrating the spatial distribution of the dimensionality of the fast subspace and the structure of the fastest and active modes. We identified the dominant processes defining these modes, and highlighted their spatial distribution. Of particular note, relevant to the fastest chemical mode (whose structure is clearly consequential to requisite time integration strategies), is the observation of the dominace of R_{204} : NNH = N₂+H everywhere inside the edge flame, with NNH being the associated CSP radical. On the other hand, a number of CH and $CH_2(S)$ reactions dominate the fastest mode in the premixed flame preheat zones around the outer rim of the edge flame, with $CH_2(S)$ being the associated CSP radial.

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