THE EFFECTS OF FLAME STRUCTURE ON EXTINCTION OF CH₄-O₂-N₂ DIFFUSION FLAMES

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The effects of flame structure on the extinction limits of CH₄-O₂-N₂ counterflow diffusion flames were investigated experimentally and numerically by varying the stoichiometric mixture fraction Zₛ. Zₛ was varied by varying free-stream concentrations, while the adiabatic flame temperature T₉₅ was held fixed by maintaining a fixed amount of nitrogen at the flame. Zₛ was varied between 0.055 (methane-air flame) and 0.78 (diluted-methane-oxygen flame). The experimental results yielded an extinction strain rate Kₑ of 375 s⁻¹ for the methane-air flame, increasing monotonically to 1042 s⁻¹ for the diluted-methane-oxygen flame. Numerical results with a 58-step C1 mechanism yielded 494 s⁻¹ and 1488 s⁻¹, respectively. The increase in Kₑ with Zₛ for a fixed T₉₅ is explained by the shift in the O₂ profile toward the region of maximum temperature and the subsequent increase in rates for chain-branching reactions. The flame temperature at extinction reached a minimum at Zₛ = 0.65, where it was 200°C lower than that of the methane-air flame. This significant increase in resistance to extinction is seen to correspond to the condition in which the OH and O production zones are centered on the location of maximum temperature.

Introduction

Studies of strained laminar diffusion flames have proven useful, both for developing a fundamental understanding of flame structure and extinction and for application in the flamelet model of turbulent combustion. Flame extinction resulting from excessive strain is of particular importance and has received considerable attention. For a strained diffusion flame, Lifian's [1] results for one-step irreversible kinetics have shown that, for a given free-stream temperature, the oxidizer or fuel can leak through the flame. Which reactant leaks through depends on the ratio of oxidizer to fuel mass fractions. In the present work, the stoichiometric mixture fraction,

\[ Zₛ = (1 + Y_{F,C} Wₐ Y_{O,C} Y_{O,-} W_{F,V} v)^{-1} \]  

is used to characterize this ratio, where \( Y \) is the mass fraction, \( W \) is the molecular weight, \( v \) is the stoichiometric coefficient, and the subscripts \( O \) and \( F \) refer to oxidizer and fuel, respectively. For \( Zₛ < 0.5 \), oxidizer leaks through, whereas for \( Zₛ > 0.5 \), fuel leaks through. A different reactant penetrates the flame with different \( Zₛ \) because with one-step kinetics, the controlling feature for reactant leakage is the global structure of the flame, which is dictated only by the composition of the free-streams (assuming the temperatures of the free-streams are equal).

For the methane-air flame (\( Zₛ = 0.055 \)), it is well-known that oxygen leaks through the flame while methane is completely consumed [2]. One-step asymptotic analysis incorrectly predicts fuel leakage because, although the basic flame structure suggests fuel leakage, the rate-limiting oxidation kinetics result in oxygen leakage. Reduced mechanisms with two steps or more can capture separately the kinetics of fuel and oxygen consumption and thus predict oxygen leakage [2]. Although Lifian's results do not correctly identify which reactant leaks through the flame, they do demonstrate that extinction is intimately linked with outer flame structure, as dictated by \( Zₛ \), such that when the basic structure of the flame is altered, the extinction strain rate will be affected.

A common example of the effects of free-stream composition on extinction is the decrease in Kₑ that occurs when reactants are diluted [3–5]. Fuel dilution decreases the flame temperature but also increases the stoichiometric mixture fraction, moving the flame away from the oxidizer and toward the fuel. The reduction in flame temperature slows chain branching while only weakly affecting radical recombination, making it easier to extinguish the flame. However, the basic structure also changes due to the shift in the flame location. To the best of the authors' knowledge, a systematic study of the effects of the shift in flame location on flame structure and extinction has not been performed.

Ishizuka and Tsuji [4] studied the effect of inert addition on extinction of methane flames. They diluted either the fuel or the air stream to determine the limiting fuel and oxidizer concentrations for stable burning. Their results showed that there is a
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TABLE 1
Extinction Conditions for CH$_4$-O$_2$-N$_2$ Flames with $T_{ad} = 2226$ K.

<table>
<thead>
<tr>
<th>Flame</th>
<th>$Z_a$</th>
<th>$Y_{CH_4}$ at $Z = 1$</th>
<th>$Y_{O_2}$ at $Z = 0$</th>
<th>$K_{ef}$ (s$^{-1}$) (exp.)</th>
<th>$K_{ef}$ (s$^{-1}$) (num.)</th>
<th>$T_{ef}$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.055</td>
<td>0.35</td>
<td>0.233</td>
<td>375 ± 10</td>
<td>494</td>
<td>1770</td>
</tr>
<tr>
<td>B</td>
<td>0.35</td>
<td>0.157</td>
<td>0.339</td>
<td>482 ± 10</td>
<td>814</td>
<td>1732</td>
</tr>
<tr>
<td>C</td>
<td>0.50</td>
<td>0.111</td>
<td>0.437</td>
<td>601 ± 10</td>
<td>996</td>
<td>1651</td>
</tr>
<tr>
<td>D</td>
<td>0.65</td>
<td>0.0845</td>
<td>0.632</td>
<td>770 ± 20</td>
<td>1226</td>
<td>1572</td>
</tr>
<tr>
<td>E</td>
<td>0.78</td>
<td>0.0709</td>
<td>1.0</td>
<td>1042 ± 20</td>
<td>1458</td>
<td>1656</td>
</tr>
</tbody>
</table>

The adiabatic flame temperature for this reaction is 2226 K. The stoichiometric ratio of CH$_4$ + 2O$_2$ + 7.52 N$_2$ → products (2)

Experimental and Numerical Methodology

The experimental apparatus is described in detail in Du and Axelbaum [6]. Briefly, a flame is established between two 11-mm opposed jets spaced 8 mm apart. The jets emanate from tubes with honeycomb cores placed 50 cm upstream of each exit. An annular co-flow of nitrogen is added to both fuel and oxidizer to eliminate oxygen entrainment from ambient air and to extinguish the flame that would exist outside the region of interest. The complete flow field has not been characterized, but it is expected to be between potential and plug flow [9].

To obtain extinction strain rates, the flow rate is increased until extinction occurs. A bypass is used to ensure that the composition remains fixed as strain rate is increased. The flame is then reestablished at conditions very near extinction, and the velocity profile along the stagnation streamline is measured with laser-Doppler velocimetry. The strain rate, $K$, is identified as the magnitude of the slope of the velocity profile upstream of the flame. Because the densities of the fuel and oxidizer free-streams are similar, the velocity gradients on both sides of the flame are identical within experimental uncertainties.

The numerical scheme employed was developed by Smooke et al. [9]. The version of the code used here assumes that the flow field outside the mixing layer can be described by potential flow. The reaction mechanism used is the a 59-step C1 mechanism given in Ref. 10.

Results and Discussion

To study the effects of flame structure, $Z_a$ must be varied while maintaining a constant adiabatic flame temperature. $Z_a$ can be varied by simply adding inert to one of the reactants. However, as noted above, this will also change the flame temperature, and in this work, the objective is to isolate the effects of flame structure resulting from shifts in flame location. Therefore, our approach is to consider flames with different values of $Z_a$ but with the same adiabatic flame temperature $T_{ad}$. To accomplish this, we start with the methane air flame as the reference condition. The stoichiometry is:

\[
\text{CH}_4 + 2\text{O}_2 + 7.52\text{N}_2 \rightarrow \text{products} \quad (2)
\]
2227 K. $Z_{av}$ can be varied without changing $T_{ext}$ simply by maintaining a fixed amount of nitrogen while varying the relative amount that is added to the fuel and oxidizer. Experimentally, this is accomplished by choosing flow rates that satisfy

$$Q_{N_2} \alpha Q_O + Q_{N_2} \alpha Q_F = 3.76$$  \hspace{1cm} (3)

where $Q_{N_2}$ is the flow rate of nitrogen mixed with the $i$th species, and subscripts O and F refer to $O_2$ and $CH_4$, respectively. To obtain the widest range of $Z_{av}$ and thus more clearly delineate structural effects, flames are considered where all the nitrogen is introduced with the oxidizer (methane air flame, $Z_{av} = 0.055$) to the other extreme where it is introduced with the fuel, such that the oxidizer is pure oxygen ($Z_{av} = 0.78$).

The validity of this approach to maintaining constant flame temperature is demonstrated in Fig. 1, where the maximum temperature obtained from the numerical calculations is plotted as a function of $1/K$ for $Z_{av} = 0.055$ and for $Z_{av} = 0.78$. As $1/K \to \infty$, that is, in the limit of infinite Damköhler number, the temperatures of both flames asymptote to a common value, which is as expected because the adiabatic flame temperatures are the same. Nonetheless, the behaviors near extinction vary considerably. For low $Z_{av}$, the flame temperature is nearly constant until very close to extinction, where it abruptly falls off. For large $Z_{av}$, the temperature falls off much more gradually and persists to a much lower temperature. Though the fuel and $T_{ext}$ are the same, the response of the flames to increasing $K$ are very different.

The experimental extinction strain rates, averaged over five separate measurements, are listed in Table 1 for five different flames, flames A-E. For the methane air flame, $K_{ext} = 375$ s$^{-1}$, which is in good agreement with the results of Chelliah et al. [11], who found $K_{ext} = 380$ s$^{-1}$. The extinction strain rates are seen to increase with $Z_{av}$, even though $T_{ext}$ is constant. For example, the strain rate required to extinguish the diluted methane-oxygen flame is almost three times as high as for the methane-air flame.

Table 1 also includes numerical results for $K_{ext}$ and the flame temperature at extinction, $T_{ext}$. In Fig. 2, experimental and numerical results for $K_{ext}$ are plotted as functions of $Z_{av}$. The numerical results are found to correctly predict the increase in $K_{ext}$ with $Z_{av}$, yet they consistently overpredict $K_{ext}$ by 30-50%. This discrepancy for the methane air flame has been observed by others and is due to the simplified kinetic scheme employed in the model and the assumption of potential flow outside the mixing layer, when the actual flow field is somewhere between potential and plug flow [11]. For the purposes of this study, a precise modeling of the flow field is not necessary and the potential flow assumption should be adequate. Thus, the qualitative agreement between experiment and theory over this wide range of $Z_{av}$ demonstrates that C2 kinetics are not needed to predict the observed trends, and that the C1 scheme employed is sufficiently valid to allow it to be used to extract an understanding of how $Z_{av}$ affects the structure of methane flames.

Before considering the results from detailed kinetics, it is instructive to evaluate how the outer structure of the flame is affected by increases in $Z_{av}$ by considering a simple flame sheet argument. Since the adiabatic flame temperatures of flames A-E are the same, the primary effect of increasing $Z_{av}$ is to shift the flame from the oxidizer toward the fuel. In doing so, the outer structure of the flame is affected.

Considering the equilibrium (flame sheet) solution in Z space, shown in Fig. 3, the changes in outer structure are readily interpreted. For purposes of discussion, let $T_{av}$ represent a temperature that would define the boundaries of the reaction zone if the kinetics were finite. Noting the temperature profiles and the nominal boundaries of the effective reaction zone, it follows that for flame A, there is, on
Now, considering flame E, which has the highest value of \( Z_T \), it is clear that the average oxygen mass fraction within the reaction zone is much higher than that of flame A. The higher mass fraction within the high-temperature region of the flame would be expected to accelerate branching chemistry and produce more radicals. Thus, the flame would be stronger and more difficult to extinguish.

The validity of the preceding interpretation is confirmed by the numerical investigations employing detailed kinetics and transport properties. Figure 4 shows the reactant and temperature profiles obtained for flames A and E, plotted in mixture fraction space to allow for convenient comparison with Fig. 3. The high strain rate curves in Fig. 4 are at their corresponding extinction limits. These results show that oxygen leakage is common to all flames, as is the absence of methane leakage. Therefore, the oxygen consumption reactions are rate limiting at high \( Z_T \), as they are for the methane air flame. Other than the increase in oxygen leakage, the methane and oxygen profiles are little affected by increasing strain rate. The temperature falls off, and the peak temperature shifts towards smaller \( Z_T \).

Figure 5 shows the profiles of temperature and
The combination of high temperature and large concentrations of OH and O are higher in flames with extinction shown for flames A and E. The concentration of H, OH, O, and temperature in the high-temperature zone increases as Zst increases. As Zst increases, the flame location moves toward the fuel boundary. With further increases in Zst, the flame location moves past the stagnation point toward the fuel boundary. In addition, the amount of oxygen in the high-temperature region increases as Zst increases. For example, at $T = 1600$ K on the oxidizer side, there is 5.7 mole % O₂ for flame A as compared to 23.0 mole % for flame E. Thus, these findings are consistent with the flame sheet arguments described previously.

The amount of fuel in the high-temperature zone is considerably greater in flame A than in flame E. Although the rate of fuel pyrolysis is not expected to have a dominant effect on flame extinction, this finding does have important implications for soot formation; and in Ref. 6, the effects of flame structure on soot formation have been discussed.

In Fig. 6, the profiles of O, H, and OH near extinction are shown for flames A and E. The concentrations of OH and O are higher in flames with higher Zst, whereas those of H are somewhat lower. The combination of high temperature and large oxygen concentration in the vicinity of the hydrogen radical pool accelerates the primary chain branching reaction,

$$
H + O_2 \rightarrow OH + O \quad (4)
$$

because this reaction has a high activation energy and is first order in O₂ concentration. Thus, higher Zst favors production of OH and O and consumption of H. Numerical results for flames with $K = 1/2K_{ref}$ have also been obtained to ensure that the given trends are not anomalies associated with the unstable conditions at extinction. Though the radical pool increases when K is decreased, the trends are consistent with those of Fig. 6 and lead to the same conclusions as to the effects of Zst.

The $Zst = 0.78$ flame can withstand a lower temperature before extinction because the increases in OH and O provide greater resistance to extinction, allowing the flame temperature at extinction $T_{ref}$ to be lower. As shown in Fig. 2, the dependence of $T_{ref}$ on Zst is not monotonic. $T_{ref}$ approaches a minimum of 1299°C near Zst = 0.65 (flame D), which is 200°C lower than for the methane-air flame. Figure 7, which is a plot of rates for the radical production reactions for flames A and D, suggests an explanation for this behavior. The reaction rates for these reactions, particularly Eq. (4), are centered at the
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location of maximum temperature for flame D, whereas they are far from this location for flame A. Thus, the temperature where the hydroxide and oxygen radicals are produced is comparable to the flame temperature for flame D, as opposed to about 100°C less for flame A. Consequently, the flame temperature can be reduced more for flame D than for flame A before the temperature in the radical production zone is reduced to an extent that the flame can no longer support combustion. For $Z_{at} > 0.65$, the radical production zone is shifted farther toward the fuel, and the maximum temperature must again be higher to sustain radical production.

Conclusions

For the CH$_4$-O$_2$-N$_2$ system at a given adiabatic flame temperature, the extinction limits increase with stoichiometric mixture fraction. This increase, which results from the shift in the O$_2$ profile into regions of higher temperature, raises the OH and O production rates and concentrations in this region, yielding stronger flames. This shift in the relative temperature and O$_2$ profiles also results in a difference of up to 200°C in the extinction temperature for the same fuel and the same adiabatic flame temperature. This result is contrary to what is suggested from the data of Ishizuka and Tsuji [4], that there is a unique limit temperature that constrains the extent of dilution that a flame can sustain. The position of the radical production zone relative to the maximum temperature appears to be critical in identifying the limit temperature for stable burning.

The results of this study are also relevant to practical flames wherein the fuel is diluted and/or the oxidizer is enriched. There are a number of applications where enriched oxygen is presently used, notably in blast furnaces. Enriched oxygen combustion has also been proposed to reduce particulates, for example, in diesel engines [12].

REFERENCES