Observation of Double Flame Structures in Near-Limit Premixed Flames

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1 Introduction

In order to make further progress in the design of advanced internal combustion engines which offer heightened efficiencies and reduced emissions, more accurate predictions of low-temperature heat release are necessary [1]. Low-temperature heat release is produced during first-stage ignition, often called the cool flame stage. While the existence of cool flames has been known for centuries [2], new and unexpected cool flame phenomena are still being discovered today. Nayagam et al. [3] found that it is possible for a large *n*-alkane droplet burning in a microgravity environment to transition into a quasisteady cool diffusion flame through radiative extinction. It has also been shown in ground-based experiments that a stable premixed hot flame can undergo strained extinction into a premixed cool flame [4]. Numerical simulations of this transition have been performed in both freely-propagating and strained geometries [5]. It was seen in these simulations that a double flame structure, in which an upstream cool flame is followed by a near-limit hot flame, can act as the transitional stage between the classic hot flame and the cool flame.

However, while it has been previously demonstrated experimentally that the capability of a hot flame to transition to a cool premixed flame can increase the flammability limits of the mixture [4, 6], the transition itself has not been studied in detail. The purpose of this investigation, therefore, is to examine the mechanism by which a near-limit premixed hot flame becomes a transitional double flame before extinguishing into a premixed cool flame. Premixed dimethyl ether (DME) flames are studied using an ozone-assisted counterflow burner.

2 Experiment

The experimental platform for this investigation consists of a counterflow burner coupled with an ozone generator [7]. A schematic of the integrated setup can be seen in Figure 1. The exit diameter of both the upper and lower burner nozzles is 1.3 cm, and the burner separation distance is set to 2.25 cm for all measurements in this study. Pure nitrogen is issued from the upper burner at a temperature of 550 K,

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which decomposes the ozone in the premixture (supplied by the lower burner) in order to ignite the cool flame. The oxidizer stream initially consists of pure oxygen, which passes through an ozone generator to produce an O_2/O_3 stream. The fuel stream is then mixed in with the oxidizer, and the DME/ O_2/O_3 premixture exits the lower burner at 300 K. DME is chosen over other fuels due to its gaseous state at room temperature (heating the premixture would prematurely decompose the ozone) and strong low-temperature chemistry.

In this study, the ozone concentration refers to the mole fraction of ozone in the oxidizer stream exiting the ozone generator. The strain rate (a) is defined as the density-weighted gradient of the axial flow velocities [4]. The equivalence ratio (ϕ) refers to the ratio of the total moles of fuel



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Figure 1. Schematic of the experimental setup. including the ozone generator and CH₂O PLIF system.

(DME) to the total moles of oxidizer $(O_2 + O_3)$ divided by the stoichiometric ratio. All measurements are taken at atmospheric pressure.

In order to visualize the premixed flame structure, planar laser-induced fluorescence (PLIF) of CH₂O molecules is performed [8]. The third harmonic (355 nm) of an Nd:YAG laser (Quantel, Q-smart 850) excites the CH₂O at an energy of approximately 200 mJ/pulse. A low-pass filter (390 nm) is positioned in front of an ICCD camera (Princeton Instruments, PI-MAX 4) to isolate the CH₂O fluorescence. The laser beam is expanded into a 200-µm-thick vertical sheet and positioned at the centerline of the burner. For chemiluminescence measurements, the filter is removed from the ICCD camera.

The numerical modeling in this study employs the AramcoMech 1.3 [9] kinetic model for DME chemistry.





Figure 2. Plots of (a) maximum chemiluminescence and (b) the location of maximum chemiluminescence in the transition from a hot premixed flame to a cool premixed flame, during which the equivalence ratio is gradually decreased from $\phi = 0.102$ to $\phi \approx 0.090$ [6].

Figure 3. False-color high-speed video images (125 fps) of the transition from a double flame to a cool premixed flame at $a = 59 \text{ s}^{-1}$ (momentum imbalanced) and 3.5% O₃. The equivalence ratio is near $\phi = 0.090$ during this sequence.

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The ozone submodel consists of the reactions proposed by Ombrello et al. [10] with the update from [11]. Calculations of steady-state species profiles are performed by the OPPDIF [12] module of the CHEMKIN package with a plug flow assumption

3 Results

For cases in which the extinction limit of the cool premixed flame exceeds the extinction limit of the hot flame, a double flame stage occurs before the hot flame is fully extinguished, as briefly mentioned in [6]. Figure 2 shows the temporal evolution of a near-limit DME hot flame extinguishing into a cool flame. As the equivalence ratio is reduced, the hot flame retreats from the bottom burner (indicating a decrease in the flame speed) and the brightness of the chemiluminescence concurrently decreases (indicating a drop in the flame heat release). For a brief period of time before hot flame. The hot flame portion of the double flame eventually extinguishes, leaving only a cool flame. The transition can be seen visually in Figure 3. The double flame contains two distinct reaction zones at t = 28.14 s. Extinction of the downstream hot flame then occurs at t = 28.15 s, resulting in a lone cool flame (t = 28.18 s).

The experimentally measured steady-state flame structures of each of these stages—the hot flame, the double flame, and the cool flame—are depicted in Figure 4. Unfiltered visible chemiluminescence and CH₂O PLIF are measured for each case and converted into plots of centerline intensity versus distance.



Figure 4. Experimental measurements of centerline visible chemiluminescence and CH₂O PLIF from a premixed DME (a) hot flame, (b) double flame, and (c) cool flame at $a = 52 \text{ s}^{-1}$ and three different equivalence ratios.

The hot flame in Figure 4a at $\phi = 0.095$ shows a fairly typical structure, with a monotonic chemiluminescence signal peaking at the location of the reaction zone and high CH₂O concentrations in the preheat zone (recall that the premixture exits from the lower burner, i.e., the right side of the plot). As the equivalence ratio decreases, a double flame is gradually formed, as can be seen in Figure 4b for at $\phi = 0.090$. Essentially, the preheat zone, governed by low-temperature chemistry, achieves a flame speed that exceeds that of the weak, near-limit hot flame. As the hot flame weakens further and further with decreasing equivalence ratio, the hot flame speed decreases more and more, and the hot flame reaction zone (driven by high-temperature chemistry) falls further and further behind the low-temperature preheat zone, which is much less sensitive to the equivalence ratio. Eventually, the heat loss in the hot flame reaction zone exceeds the heat production, and the hot flame extinguishes, leaving behind the lone cool flame in Figure 4c at $\phi = 0.0875$. The cool flame shows a substantially broader CH₂O PLIF signal since formaldehyde is one of the primary products of the cool flame (and not simply an intermediate species). Due the lower activation energies required to sustain the cool flame, the cool flame does not extinguish until a much lower equivalence ratio (near $\phi = 0.0725$).

Complementary numerical calculations for this near-limit transition can be seen in Figure 5. Flame profiles are presented for four equivalence ratios. From the heat release rate profiles in Figure 4b, it appears that the distinction between the hot flame and the double flame is fairly arbitrary. All three of the



Figure 5. Spatial profiles of (a) temperature, (b) heat release rate, (c) DME mole fraction, and (d) OH radical mole fraction for premixed DME/O₂/O₃ flames at 4 different equivalence ratios during the transition from a hot flame to a cool flame.

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highest equivalence ratios exhibit non-monotonic heat release rate profiles. For $\phi = 0.1169$, the peak heat release in the preheat zone is substantially smaller than that of the main reaction zone, but the two are approximately equal in the $\phi = 0.1000$ case, and the preheat zone clearly dominates in the $\phi = 0.0854$ case. For the purposes of this paper, the first two cases will be classified as premixed "hot flames" (the peak high-temperature heat release is greater than the peak low-temperature heat release) and the $\phi = 0.0854$ case will be considered a premixed "double flame" (greater peak low-temperature heat release, leading to substantial separation between the preheat zone and the main reaction zone). The lowest equivalence ratio case experiences a monotonic heat release rate profile driven by low-temperature chemistry and thus qualifies as a premixed "cool flame."

The double flame at $\phi = 0.0854$ shows several other differences from its hot flame counterparts. Figure 5a indicates that its maximum flame temperature (1340 K) is substantially lower than the hot flames, which is due to the close proximity of the double flame to the hot extinction limit. It can also be seen in Figure 5c that the double flame experiences an inflection point in its fuel concentration profile, elucidating the two-stage ignition behavior of DME. The hot flames, which are not nearly as thick as the double flame, show rapid fuel consumption in a tightly coupled flame structure. The cool flame, moreover, displays only a single fuel consumption stage with massive fuel leakage into the stagnation plane.

4 Conclusion

The extinction of a near-limit premixed hot flame into a stable cool flame has been experimentally and numerically investigated for $DME/O_2/O_3$ mixtures in a counterflow burner. It is seen that the hot flame gradually transitions into a double flame as the flame speed of the low-temperature preheat zone exceeds that of the high-temperature reaction zone. The preheat zone eventually becomes an independent cool flame when the hot flame extinction limit is surpassed and the main reaction zone disappears. While this phenomenon has been verified in this study for atmospheric pressure premixed flames supported by ozone addition, future experiments should examine the transition between hot flames and cool flames without the assistance of ozone through the use of elevated pressures.

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